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Symposium on tektites

Foreword

Interest in tektites has increased tremendously during the past few years and studies concerning them have multiplied manyfold. Most of these concern, directly or indirectly, the origin of tektites, but the approaches are various, having to do both with experimental determination of compositions and properties of tektites and theoretical studies concerning possible modes of origin.

Correspondence among interested workers in the field in the spring of 1957 led to the organization of a Symposium on Tektites, which was held in Washington, D.C., on 17 and 18 June of that year, under the auspices of the National Academy of Sciences and the National Research Council. Dr. William R. Thurston, Executive Secretary of the Division of Earth Sciences, and other members of the staff gave a great deal of aid in arranging the conference.

The National Science Foundation made a modest grant that defrayed the expenses of conducting the meeting, but it was not possible to pay travel expenses of participants. Therefore, although most of the tektite workers in the United States were able to attend the sessions through the co-operation of their respective institutions, students of the problem from other countries were conspicuously absent.

Nevertheless, progress in most of the major lines of attack on the tektite problem was reported, and the meeting was successful in focusing attention on the general aspect of the problem and in generating further research. A summary report of the conference appeared in *Science* **127**, 91, 1958. Most of the papers that were presented at the meeting appear in the following pages of this special issue of *Geochimica et Cosmochimica Acta* "Symposium on Tektites," although a few of them have appeared, or will appear, in other journals. For example, the one by H. C. Urey on the

"Origin of tektites" was published in *Nature, Lond.* **179,** 556–557, 1957, and the one by J. A. S. ADAMS on "Uranium contents and alpha particle activities of tektites" will appear in the publications of the XX International Geological Congress. The Congress met in Mexico City in September 1956, but this part of the publications has not yet been distributed.

IRVING FRIEDMAN
Special Editor of the
Symposium on tektites

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Some considerations on tektites

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Abstract—Recent research on tektites in the Soviet Union has been devoted to studies of viscosities, major and minor element content, and ages by the potassium–argon method. The author reviews this work, and speculates on the probable origin of tektites. SPENCER's impactite glass theory is favoured, as explaining the major surface features and composition of tektites.

ALTHOUGH the area of the Soviet Union is very large, up to now no tektite or similar glass, so-called silica-glass, has been found. However, attempts have been made to discover and collect such material by calling these glasses to the attention of different scientific workers, especially geologists, geographers and others who systematically carry on field work. The Committee on Meteorites of the Academy of Sciences of the U.S.S.R. (Moscow), which is the central scientific institution on meteorites in the U.S.S.R., selected from its collection of meteorites standard collections of tektites, which were sent to several local Committees on Meteorites (the Urals, Central Asia, etc.). In accordance with the instructions of the now deceased Academician V. I. VERNADSKY, the former Chairman of the Committee on Meteorites, who devoted much attention to tektites, the author of the present paper prepared and published a special review on tektites in the magazine *Priroda* (Nature) in 1946 (E. L. KRINOV, 1946). Up till then no special articles on tektites or silica-glass had been published in Russian scientific literature; they were mentioned only incidentally in papers on meteorites.

For a long time tektites had been considered, and are even now considered by some scientists, to be a special class of glass meteorites. Therefore, the numerous collections of tektites which can be found in many museums of the world are kept together with meteorites. Tektites were studied mainly by specialists in meteoritics, because of their supposed meteoritic nature.

In the Soviet Union the main collection of tektites and silica-glass is kept with the collection of meteorites of the above-mentioned committee. It includes several hundreds of specimens of moldavites, several tens of specimens of indochinites and also philippinites (rizalites), one specimen of tasmanite, silica-glass from the meteoritic craters of Wabar and Henbury, and Libyan Desert glass.

RECENT SOVIET RESEARCH

In recent years Soviet investigators have made the following studies of tektites.

In 1941 Professor M. P. VOLAROVICH and A. A. LEONTYEVA published their results on the study of the viscosity of molten stony meteorites and tektites (VOLAROVICH and LEONTYEVA, 1941). The viscosity of the latter was investigated by the stretch–pivot method, which was worked out by the above-mentioned authors for determining the viscosity of quartz glass (VOLAROVICH and LEONTYEVA, 1936) and also obsidians (VOLAROVICH and LEONTYEVA, 1939) in connexion with the process of formation of pumice. It enables the viscosity within the limits

$10^{7.5}$–10^{10} P to be determined. This corresponds to an interval of temperature 950°–1150°C. For studying the viscosity at higher temperatures (up to 1400°C) a method based on Stokes law was applied. The above investigators had used it formerly for measuring the viscosity of boric anhydride (VOLAROVICH and LEONTYEVA, 1939) and the system B_2O_3–SiO_2 in the melted state.

The viscosity of two tektite specimens, a moldavite and an indochinite, and also one specimen of the Saratov stony meteorite (chondrite) was determined. Measurements by the stretch–pivot method were repeated, and are plotted as dots and circles in Fig. 1. Stokes law measurements are plotted as crosses. The points

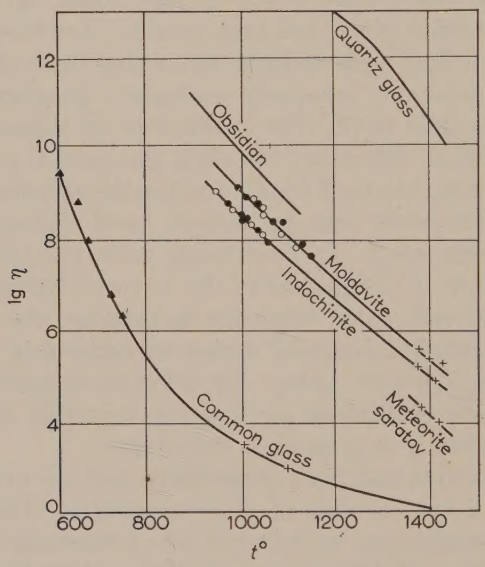

Fig. 1. Log of the viscosity of various glasses versus temperature.

for each specimen can be joined by one curve, which is very close to a straight line. The viscosity of tektites is seen to lie between that of quartz and ordinary glass, that of obsidian is somewhat larger, and that of the meteorite is very near to that of tektites.

In discussing their results the investigators, because of insufficient data, did not come to any final conclusions on the nature of tektites and pointed out the necessity of further special studies.

The second paper dealing with tektites (MALYOUGA, 1949) did not consider in detail the composition of tektites from the point of view of their origin and states that it is impossible to decide the question in such a manner. Nevertheless, in the beginning of the paper he notes that tektites are more nearly similar in TiO_2 and Al_2O_3 content to the lithosphere than to meteorites. He considers this to be one of the arguments against the extra-terrestrial origin of tektites. In order to solve

the tektite problem MALYOUGA investigated them by a method of a genetic estimation of the quantitative ratios of elements of the iron family on the basis of the law of the inconsistency of ratios in paragenetic series. As was pointed out by MALYOUGA, it is possible to apply this method only if there are sufficiently exact data on the content of such elements as vanadium, chromium, nickel, cobalt,

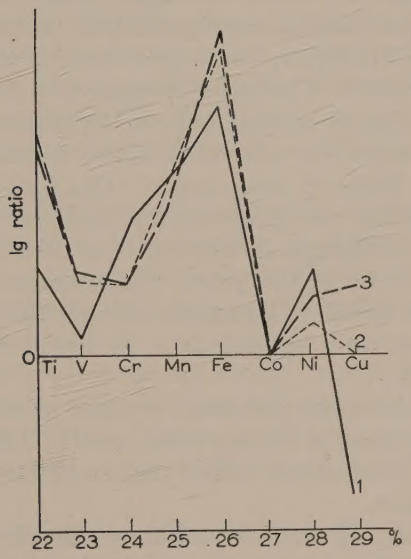

Fig. 2. Log of the ratio of various elements to cobalt for (1) stony meteorites, (2) tektites and (3) sedimentary rocks.

copper, titanium manganese and iron. The concentration of the main elements of the family Fe, Ti, Mn is quite constant in all tektites, irrespective of the place where they were found. However, there are sometimes sharp deviations in the concentration of the other elements of the family, for instance chromium and nickel. It is especially interesting, the author points out, that the indochinites of southern and northern Indo-China differ from each other by their nickel and chromium content in a greater degree than, for example, australites and the tektites of northern Indo-China.

Using data on the composition of tektites, published by PREUSS in 1935 and in addition determining cobalt as well as checking the content of other elements, MALYOUGA found the following mean distribution of elements of the iron family in tektites: 0·42 per cent Ti, 0·0068 per cent V, 0·0067 per cent Cr, 0·078 per cent Mn, 3·08 per cent Fe, 0·0009 per cent Co, 0·00239 per cent Ni, 0·0008 per cent Cu, which corresponds to the following ratios, if the Co content is taken as unity: 466 : 7·5 : 7 : 86 : 3422 : 1 : 2·5 : 1. The comparison of these data with the corresponding data for sedimentary rocks shows a close resemblance. However, there is a sharp difference from the respective data on stony meteorites, as shown

on the graph (Fig. 2). MALYOUGA concludes that tektites are not of extra-terrestrial origin. He also refutes SPENCER's hypothesis (see further), in which the formation of tektites is attributed to explosive impact of gigantic meteorites during which terrestrial rocks and quartz sand melt to form glassy masses.

The third paper deals with the determination of tektite ages by the argon–potassium method. Study by GERLING and YASCHENKO (1952) and in more detail by YASCHENKO and GERLING 1953. In order to calculate the age by the above method it is necessary to know the argon as well as the potassium content of the specimen. The argon content was determined with an apparatus constructed by HLOPIN and GERLING (1932), by prolonged heating of a pounded specimen of tektite in a vacuum at 1250°C. The latter was put into a quartz tube, joined with the help of a coupling to the measuring part of the instrument. All of the tektite melted and the emitted argon was separated in the ordinary way from other gases and measured with the McLeod manometer. The following tektites were used for the investigation: moldavites, indochinites, and philippinites (rizalites). As a result the following data on the age of tektites were derived: moldavites $< 3{\cdot}1 \times 10^6$ years, indochinites $< 4{\cdot}6 \times 10^6$ years, philippinites (rizalites) $< 1{\cdot}2 \times 10^7$ years. The tektites were found to be much younger than stony meteorites. The age of the latter, derived in a similar way, is 650×10^6 to 4500×10^6 years (GERLING and RIK, 1956).

SUESS, HAYDEN and INGHRAM published a paper in 1951 on the age of tektites determined by the argon method (SUESS *et al.*, 1951). The age was found to be of the same order, namely: australites $< 10 \times 10^6$ and 32×10^6 years, philippinites (rizalites) $< 73 \times 10^6$ years.

In conclusion GERLING and YASCHENKO state that the almost total absence of argon and the large potassium content raises doubt as to the extra-terrestrial origin of tektites, as their age is not more than that of the rocks where they were found. In their second, more extensive article YASCHENKO and GERLING (1953) considered in detail different hypotheses suggested by various authors on the origin of tektites. In conclusion they point out that the problem of the origin of tektites has not been solved as yet with any degree of certainty.

In giving the results of tektite investigations made in the Soviet Union the author of the present paper did not have as his aim a critical review. He would only like to note that all three studies, which were made independently, deal with three different aspects of tektites; in one the extra-terrestrial origin of tektites is decidedly refuted; in the other two the question is left open.

In addition to this hypothesis there is another, that tektites are a special class of glass meteorite, which is supported and persistently defended by a group of investigators (CASSIDY, 1956).

The comparison of tektites with other naturally occurring glasses and reviews of theories of their origin have been made by various workers and need not be repeated here (HEIDE, 1938, 1939; BARNES, 1940; OSWALD, 1942; FENNER, 1949; BAKER, 1956). Of the many origins proposed, the meteorite impact theory of SPENCER (1933) deserves the most attention. SPENCER considers the formation of tektites analogous to that of silica-glass from known meteoritic craters, i.e. that

they are the product of melting of terrestrial rocks, caused by the explosive impact of gigantic meteorites during the formation of meteoritic craters.

Let us examine the main facts which speak in favour of SPENCER's hypothesis on the origin of tektites.

Many meteoritic craters are present in various places on the earth's surface. The structure of the craters and the presence of scattered small fragments of iron meteorites around them on the one hand, and on the other hand theoretical investigations of the fall of gigantic meteorites, which after passing through the atmosphere retain a cosmic velocity, show convincingly that the fall of such meteorites is accompanied by an explosion. As a result of the explosion the main mass (if not all) of the meteorite evaporates, around the place of fall a negligible number (in comparison to the whole mass) of small fragments of the meteorite are scattered and at the place of fall—explosion depressions—meteoritic craters are formed.

The presence of masses of the so-called silica-glass around and in each meteoritic crater of explosive origin is a characteristic peculiarity of these craters. The presence of pieces of silica-glass confirms the explosive character of the formation of such craters and also shows that during the explosion of gigantic meteorites terrestrial rocks are melted to such a degree that masses of glass, silica-glass, are formed. The latter observation is especially convincingly confirmed by the discovery in the silica-glass from the Henbury and Wabar meteoritic craters of tiny iron–nickel globules. These globules are hardened tiny drops, which were splashed during the explosion of the meteorite and caught by the melted glass masses (SPENCER, 1933).

All tektites, irrespective of the place of their discovery, show a remarkable similarity of form and type of surface structure. The main and characteristic forms of tektites are the following: spherical, ellipsoidal-flattened, sand-clocks or dumb-bells, flat plates with rounded edges, drop-like forms, and others. Typical elements of the surface structure among others are: spherical or ellipsoidal pits of various dimensions, worm-like grooves, rice-shaped small depressions, and furrows which are either curved or spread fan-like over the surface. The fact that the above forms and types of surface structure of tektites are repeatedly met with in all places of discovery is really striking.

The difference between tektites discovered in various places is mainly a difference in colour observed in transmitted light. For example, moldavites are green and indochinites brown. Fewer bubbles are in tektites than in silica-glass, therefore tektites are denser and more stable than silica-glass. However, the greatest difference between tektites and silica-glass is in their form. As was mentioned above, tektites have definite shapes and surface structure, which shows that there is a common cause for the formation of these types of form and surface structure. Silica-glass on the other hand is represented by pieces of indefinite shape, the surface being characterized by numerous spheroidal, ellipsoidal and elongate pits, resulting in a bubbled or slag-like appearance.

This sharp difference of form and surface structure of tektites and silica-glass is observed only when comparing typical representatives of each group. However, there are specimens among tektites, as well as among silica-glass, which are

transitional types. For example, in the collection of the Committee on Meteorites, among many typical specimens of indochinite, there is one 9·6 × 6·6 × 3·1 cm weighing162 g, which has no definite form and in this respect strongly resembles a piece of silica-glass. Besides its nondescript shape it also has a bubbled surface similar to silica-glass. This specimen for some reason eluded shaping to which other specimens of indochinites were subjected.

An especially distinct transitional type is found among pieces of silica-glass from the Libyan desert. The latter, according to their density and stability are entirely similar to tektites. They also have exactly the same spherical and ellipsoidal pits on their surfaces as are observed on the surfaces of typical tektites. However, these pieces, similar to the above-mentioned specimen of indochinite, are formless, although their surfaces, edges and corners are smooth.

Many investigators, following Suess, consider tektites to be glassy meteorites and that the form and surface structure of typical tektites are a result of the action of the atmosphere on the tektites during their flight at cosmic velocity in the earth's atmosphere.

The author (Krinov, 1955), who has devoted much time to the study of the morphological properties of iron and stony meteorites, including the form, surface relief and surface structure of the fusion crust, decidedly opposes such a conclusion. In reality there are no traces, even the smallest, of the action of the atmosphere on tektites, their form, surface relief, or structure.

The form and surface structure of tektites decisively show that they are the result of mechanical and chemical erosion. We can therefore regard tektites as a kind of pebble of silica-glass.

The absence of typical forms and surface structure of silica-glass from meteoritic craters similar to those which are observed in tektites is explained, in the author's opinion, by the considerably later date of formation of the meteoritic craters, in consequence of which both craters and pieces of silica-glass are preserved. The absence of typical tektite forms on pieces of silica-glass from the Libyan desert can evidently be explained by the character of mechanical and chemical erosion in a dry climate, which differs somewhat from that where other tektites are found. No crater has been found in the vicinity of the Libyan Desert glass; perhaps denudation destroyed it. A careful and detailed study of the surface structure of tektites shows that the tektites of each separate group have their own specific peculiarities of surface structure. These peculiarities could have been caused by some differences in the character of erosion.

Conclusions

On the basis of the above facts and conclusions, the author makes the following supposition about the origin of tektites.

Tektites are glassy objects with an origin similar to that of silica-glass from meteoritic craters; consequently tektites are fusion products of terrestrial rocks resulting from explosions upon the impact of gigantic meteorites.

One can surmise that in the far past, during a long period of the history of the earth's crust, a large number of meteorites fell on the surface of the earth accompanied by explosions. During this period practically all the surface of the earth

was probably covered by such meteoritic falls and the attendant meteoritic craters and silica-glass. Because of the different composition and character of the material in which the meteoritic craters formed in various parts of the earth, masses of silica-glass with different stability originated.

In the course of time most of the meteoritic craters were destroyed, and those preserved were the youngest and those on firm stony ground. Only in such craters were fragments preserved of iron meteorites and the relatively less stable pieces of silica-glass.

The overwhelming majority of meteoritic craters and the meteoritic fragments about them have long since disappeared. The craters were destroyed by denudation and the fragments by weathering. Only the most stable pieces of silica-glass, the so-called tektites, have been preserved being silent witnesses of the fall in the far past of gigantic meteorites. Under the influence of various atmospheric and soil agents the tektites were moved and scattered over the earth's surface during the very long period of time which elapsed since they were formed. One must however bear in mind that the tektites discovered in each region could have formed as the result not of one meteoritic fall but of many. For instance the tektites in Australia could have been formed by the fall of many meteorites at different times and places over much of the continent.

In order to solve the problem of the validity of SPENCER's hypothesis on the origin of tektites the author considers that experiments for making artificial tektites are necessary. Various initial materials from different regions of the earth should be used and various conditions for the formation of tektites (temperature, pressure, etc.) provided. In the author's opinion such experiments will either finally confirm or refute SPENCER's hypothesis.

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Properties of tektites pertinent to their origin*

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Abstract—Theories of the origin of tektites are examined to see if they explain the following critical properties: chemical properties, lechatelierite particles, two periods of melting, flow structure, shapes and sizes, and distribution of age.

A volcanic origin either from the earth or the moon cannot explain the above properties; neither can an origin from lightning, or as impactites from the moon, or from collisions of asteroids with each other.

The impact theory of Urey can explain the tektites if the molten rock was thrown beyond the earth's atmosphere and was remelted upon its return. The absence of partially fused tektites is against such a hypothesis.

The formation of tektites by the destruction of a planet similar to the earth should not be overlooked.

Introduction

The various occurrences of tektites and other sporadic glasses have been adequately described in many excellent papers and treatises and their description need not be repeated here. A bibliography on tektites (Barnes, 1940) is essentially complete; for the ensuing 10 years papers on meteorites including tektites are listed by Brown (1953). For easy reference, important publications reporting new occurrences and descriptions of various groups are given in an appendix.

To be acceptable, any theory of origin of tektites must explain their distribution and all their properties, some of which are more closely bound to their origin than others. The critical properties are listed as follows:

(1) Chemical composition.
(2) Lechatelierite particles.
(3) Two periods of melting.
(4) Flow structure.
(5) Shapes and sizes.
(6) Distribution and age.

Each of these characteristics is examined in the following pages to see if it can be acquired only through a terrestrial origin, only through an extra-terrestrial origin, or if it can be acquired through either. Any property that can result from an extra-terrestrial origin will be further examined herein to see if evidence points to any one type of parent body as the most likely source.

Any valid theory of origin for tektites must fit the available evidence. Direct evidence such as that furnished by the tektites carries the most weight. Evidence obtained from comparison with accessible materials is only on a slightly less secure footing. Indirect evidence such as that furnished by astronomical observations is the least secure because only a few properties are measured. However, any theory of extra-terrestrial origin of tektites must not transcend the laws of motion—the field of the astronomer and mathematician rather than of the

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geologist and geochemist. Likewise, any theory that does not satisfy all the geological and chemical evidence will founder. Because there has been a strong tendency for critical geological evidence to be ignored during the past 15 years, all geological evidence that must be satisfied in proposing an origin for tektites is reiterated in this paper.

Critical Tektite Properties

Chemical composition

All available chemical analyses of tektites, sixty-eight in number, were assembled by Barnes (1940, p. 524–532) and the data critically examined (p. 533–543). It was shown that tektites mostly are closely allied in composition to argillaceous sedimentary rocks, and that out of thousands of analyses of igneous rocks only a very few compare closely to tektites. Linck (1928) may have been the first person to notice the similarity in composition of tektites and sedimentary rocks. Silverman (1951) investigated the isotope geology of oxygen and found that the higher isotope ratios for tektites bring them within the range of sedimentary rocks. Adams (1956) found the uranium concentration in tektites also to be similar to that in sedimentary rocks. However, the most recent analysis of a bediasite (Barnes, 1956) definitely shows it to have acid igneous characteristics, as also do tektites from the Ivory Coast.

The chemical composition of tektites may not appear significant to non-geologists because they probably are not familiar with the great volume of research on the composition of the major groups of earthly rocks which has been so ably synthesized by Clarke (1924). Igneous rocks were the first rocks at the surface of the earth, and the first sedimentary rocks were of necessity derived from igneous rocks. As time passed, metamorphic rocks produced from both igneous and sedimentary rocks came into prominence, sedimentary rocks were folded into mountain chains and otherwise uplifted, and all groups of rocks thereafter contributed material to the sedimentary sequence. Certain definite chemical changes took place during the weathering and sedimentational processes with the result that the chemical composition of sedimentary rocks is mostly distinct from the chemical composition of igneous rocks.

Sedimentary rocks could not have formed on the moon because there is no hydrosphere nor atmosphere. From this viewpoint alone it seems very unlikely that the moon could be the source of tektites. Also, by analogy with the manner in which earthly igneous rocks have evolved, it is believed that rock as acid as tektites is unlikely to be present on the surface of the moon.

Earthly igneous rocks have been in the process of evolving throughout the earth's history, with segregation of less dense materials, until now large areas of continents are surfaced by acid igneous rocks. It is believed that only a minimum of segregation could have taken place on the moon because of its mass (only 0·01228 times that of the earth) causing the force of gravity at its surface to be only one-sixth that at the earth's surface.

The moon's small size makes it likely that temperatures at which rocks could melt were confined to a rather short period of its history, and rocks which did melt were acted upon by a much smaller force of gravity than on earth; the

tendency for segregation, therefore, was much less than on earth. If this reasoning is correct, then the rocks at the surface of the moon should be little segregated and therefore should have a density near that of the moon as a whole (3·36); this figure is slightly in excess of the density of the more basic rocks at the earth's surface. Material of this high density could not possibly furnish glass of the low density of tektites. The density, as well as the refractive index of a glass, is directly related to its chemical composition—the higher these values the more basic the composition and conversely the lower these values the more acid the composition.

From their chemistry the logical place for tektites to have formed is on earth. The only other possibility appears to be their formation from sedimentary rocks on another planet which had a similar history to that of the earth.

Glass formed by meteorite impact, as pointed out by SPENCER (1933), contains appreciable nickel introduced from melted meteoritic material. Of the eleven tektites analysed for nickel, nickel was absent in six, present as a trace in three, and in two australites amounted to about 235 and 470 p.p.m. respectively. These amounts are within the range found in terrestrial sedimentary rocks. GOLDSCHMIDT (1937) estimated that silicate meteorites average about 3300 p.p.m. nickel. Asteroids are probably similar in composition to meteorites, and if this is true at least a few tektites should be found which are richer in nickel than any so far analysed. The data are few and possibly not representative. The origin of tektites by meteorite or asteroid impact can not be properly evaluated until many times the number of nickel analyses now available have been made on all tektite groups.

The low water content of tektites, according to FRIEDMAN (1955), sets them apart from obsidian even though the deuterium–hydrogen ratio of the two is approximately the same. During a tektite conference in Washington, D.C., 17–18 June 1957, and in a letter of 12 November 1957, FRIEDMAN pointed out "that rock melted by the first atom bomb has a water content approaching that of tektites." So far as the writer is aware, the water content of other fused sedimentary rock has not been determined, and until this is done speculation on the low water content of tektites is fruitless. For the results to be comparable, the sedimentary rock would have to be fused at a temperature above the melting point of quartz.

Lechatelierite

Sand- and silt-size glass particles were identified from their refractive index in bediasites, australites, indochinites, and moldavites by BARNES (1940, p. 505) to be lechatelierite (fused quartz). Previously these particles had been seen, but not identified, in glass formed on mountain peaks by lightning (RUTLEY, 1885); in moldavites (RUTLEY, 1885; SUESS, 1900); and in indochinites (LACROIX, 1935). BAKER (1944) illustrated these particles from both rims and central bodies of australites and also mentioned their presence in Darwin glass, Pelee's hair, and fulgurites. Similar fused quartz particles were identified in glass fused from Eocene shale in a carbon arc (BARNES, 1940, p. 512) and in glass formed by broken power lines arcing through the soil (BARNES, 1940, p. 511). ROSS (1948)

described fused quartz in the products of the first atomic bomb exploded at Alamogordo, New Mexico. The writer has a specimen of this green atomic bomb glass which shows drawn out and contorted lechatelierite (?) particles (Plate I, Fig. A) entirely similar to those found in tektites, especially the moldavites (Plate I, Fig. B).

The presence of lechatelierite in tektites furnishes critical evidence the true importance of which many students of tektites have failed to recognize; only one attempt has been made to explain them other than the explanation proposed by the writer (Barnes, 1940) that they are fused quartz grains. No theory of origin of tektites can be accepted unless it explains the presence of these particles.

Cassidy and Segnit (1955) and Cassidy (1956) attempted to explain the presence of lechatelierite by liquid immiscibility. To test for liquid immiscibility, part of specimen 34 (Barnes, 1940, p. 499), a specimen exceptionally rich in lechatelierite (Plate I, Fig. C), was heated* to 1605°C in a platinum crucible and maintained at this temperature for 30 min. About half of the lechatelierite particles disappeared, and the rest were distinctly smaller. Since this experiment was not entirely conclusive another portion of specimen 34 was maintained at 1593°C for 4 hr, and during this treatment almost all of the lechatelierite particles disappeared. This shows that these particles are not the result of liquid immiscibility and substantiates the conclusion of Barnes (1940, p. 506) that the presence of these particles is not evidence of true immiscibility; but that it is evidence that tektites were melted only a very short time.

This experiment shows conclusively that tektites could not have been derived from pre-existing glassy rock as suggested by Cassidy (1956), because any pre-existing glassy rock would have been melted sufficiently long to be thoroughly mixed; any lechatelierite particles that might have been present would have gone into solution as happened in the above experiment.

The presence of lechatelierite shows that the temperature at which tektites melted exceeded 1710°C, a fact overlooked by Houziaux (1956) who thought that a temperature of from 1200°C to 1400°C was sufficient for their formation.

The origin of tektites is limited, therefore, to some sudden manner of formation from previously non-glassy material—a material containing small quartz particles. It is unlikely that material of this composition containing quartz crystals could be present on the moon; however, material of this type is abundant on earth and presumably would be abundant on any other planet that had the same history as the earth.

Two periods of melting

The most persuasive evidence of extra-terrestrial origin of tektites is furnished by the australite button. The two periods of melting, shown in the button form, are almost incontrovertible evidence that tektites are glass objects that plunged into the earth's atmosphere. Such an object plunging into the atmosphere, if of proper shape, should melt on the forward side, the point of greatest friction. The first melt products would be stripped, but as the object slowed, some glass

* Dr. E. Joseph Weiss and Mr. John Dexter, Department of Ceramic Engineering, The University of Texas, did these experiments.

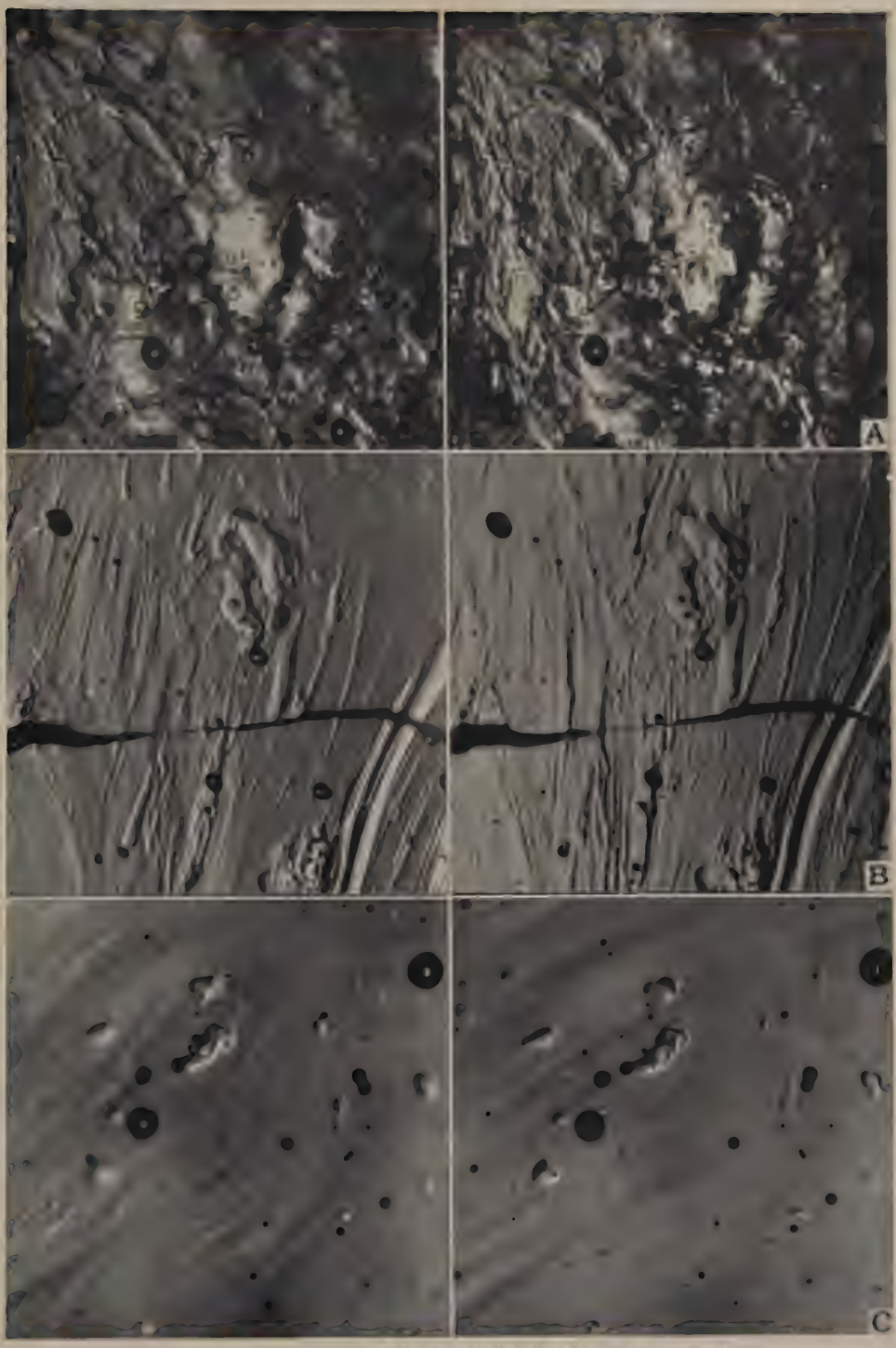

PLATE I. Stereograms* showing lechatelierite in glass. ×22.

Fig. A. Atomic bomb glass, Alamogordo, New Mexico, showing lechatelierite(?) mostly in threads, some of which are contorted. Fig. B. Moldavite (30891A), near Budweiss, southern Bohemia, showing lechatelierite in distorted grains and in threads, many of which are steeply inclined, and others which are contorted. Fig. C. Bediasite (30246–34), Grimes County, Texas, showing slightly elongate to equidimensional lechatelierite particles.

* Stereograms usually are of surfaces of opaque objects illuminated by incident light. These are of transparent objects in a transparent medium illuminated by transmitted light; unless the viewer is aware of this it may be difficult for him to see the stereoscopic relief. Bubbles are black in part with centres of white; the east–west black streak in Fig. B is a steeply inclined crack. The glass plates from which these photographs were made are 1·5 mm, 0·7 mm, and 1·4 mm in thickness respectively, thus causing the stereograms to have depth as if one were looking into transparent blocks of ice.

would flow towards the lee and curl to form a flange. Such is the form of the button as illustrated by DUNN (1908), FENNER (1934, 1935), and BAKER (1944). That most tektites do not have the button shape might be explained by the original objects not being symmetrical, causing them to tumble as they hit the atmosphere and to develop more or less equally distributed thin skins of re-fused glass.

The critical evidence furnished by the button form is that tektites were glass before they entered or re-entered the earth's atmosphere. No natural way is known to produce the button shape by terrestrial means, and even if such a shape were to form, for example by lightning, only one period of melting would be involved. The only other heat source observed on earth sufficient to melt material of tektite composition is from impact of large meteorites. Again if the glass formed by impact assumed a button shape, the second period of melting, as in the preceding example, would be absent; unless, as suggested by UREY (1955), an asteroid a mile or two in diameter plunged to earth with "energy sufficient to . . . melt some 2000 km^3 of rock." In such a catastrophe the explosive energy might have been sufficient to propel the resultant molten material beyond the atmosphere, and when solidified it would have plunged back into the atmosphere to be partly remelted.

The size of known tektite-strewn fields might be better explained by impact explosions of the correct magnitude than by any other suggested origin. Each of the half-dozen or so strewn fields would require a separate explosive impact, and the craters formed in each case should be enormous. Evidence for at least one of the half-dozen or more craters, if they ever existed, should have been found unless they all formed in areas now under water or are hidden from view by a cover of younger rock. Whether or not the mechanics involved are possible is not in the writer's field, but chemical evidence, as mentioned earlier, suggests that this did not happen.

If UREY's (1957) comet origin is correct, then the objection based on chemical composition may be invalid. However, no impact theory—either comet, asteroid or meteorite—explains the absence of partly fused material in any tektite yet examined.

Flow structure

All tektites have flow structure, as is amply illustrated by DUNN (1912), BARNES (1940), and BAKER (1944). Contorted flow structure of the type found in tektites is likely to develop only in small masses which cool quickly because of their small size. Large masses of glass, such as obsidian, cool slowly, and any flow structure retained is usually plane or gently curving and made visible by the presence of crystallites. Tektites do not contain crystallites, and the flow structure is made visible by variations in the index of refraction of adjacent layers of glass. The reason for this variation is that the chemical compositions of adjacent layers are different. Additional evidence that adjacent layers differ in composition is furnished by the severe strain which can be seen when tektite plates are viewed in polarized light. The strain is caused by adjacent layers having different coefficients of expansion. Flow structure of this type could not be expected to be retained more than a few minutes in a molten material.

That tektites were molten no more than a few minutes is proven by the experiment with tektite 34. Within 30 min after this specimen passed its melting temperature, flow structure was essentially obliterated and the resulting glass was without strain; and after 4 hr the glass was entirely homogeneous except for an insignificant number of tiny lechatelierite particles.

Any postulated origin of tektites must account for this very short period during which tektites were molten. Glass having such flow structure could be formed by meteorite or asteroid impact on the earth, moon, or some other planet, but as mentioned earlier, rock of the right composition to form such a glass could be present only on the earth or on another planet having a history similar to that of the earth.

Shapes and sizes

Much has been written about the shapes of tektites, especially of the australites (Dunn, 1908, 1912; Summers, 1913; Fenner, 1935; Baker, 1944, 1946, 1956), the indochinites (Lacroix, 1932), and the philippinites (Beyer, 1955). Even more has been written on surface sculpture, which, along with spalling, has strongly modified the shapes of the older groups. A discussion of surface sculpture and spalling is mostly irrelevant so far as the origin of tektites is concerned, because these are features acquired through etching and other erosive agencies after they came to rest. Etching, however, is helpful in revealing contorted flow structure so that tektites can be readily distinguished by superficial examination from similarly sculptured material such as obsidian.

The one form, the australite button, which appears to be inextricably related to one method of formation has been discussed above. Most of the rest of the shapes do not need any special conditions for their formation, and might well develop from spurts or splashes of glass produced in almost any manner. A few shapes found in Indo-China, such as the curved tear-drops and thin plates, however, need to be examined closely in any theory of tektite origin.

The curved, elongate tear-drop, many of which are illustrated by Lacroix (1932), is an illogical form to be produced by any of the more violent origins suggested. This form might come from glass spurted in an arc at low velocity, cooling as it went, but to have such a form developed during catastrophic explosions seems extremely unlikely. The thin plates, some with centres thinner than their edges, are also an illogical shape to be produced at supersonic speeds. These could possibly have formed from viscous masses of glass which landed on a flat surface and then spread out, but this does not explain the plates which are thinner in the centre.

A tektite mass 4 in. in length and weighing 184 g from Muong Nong, Lower Laos, has a ropy, slaggy appearance on one surface somewhat similar to that of impact glasses; however, etching is pronounced on this specimen and this observation may not be valid.

The size of individual tektites is probably critical in any evaluation of their origin. If they were obtained from pre-existing glassy rock, then fragments of glass of the size and shape of ordinary meteorites should be common. Baker (1956, p. 139) believed that australites are modifications of primary forms that

existed at the time they plunged into the atmosphere, and the writer is fully in agreement.

Libyan Desert glass fragments (Clayton and Spencer, 1934) attain weights of as much as 10 lb, whereas Fenner (1934, p. 67) found that 1993 specimens of australites averaged 0·931 g. Because of their more basic composition, australites should be more fluid at comparable melting temperatures than the high-silica Libyan Desert glass; if the two formed at about the same temperature, this might explain the difference in size of individuals in the two groups.

The indochinites are of about the same order of basicity as the australites, yet their average size is greater, indicating perhaps lower temperature of formation. The relation of the size of a tektite to the magnitude of the force creating it has not been considered so far as the writer is aware, but it seems logical that the greater the energy expended, the higher the temperature, the greater the fluidity, and consequently the smaller the tektites.

If tektites were produced by the destruction of a planet similar to the earth, as suggested by Linck (1928) and Barnes (1951, 1957), tektite sizes could easily be explained on the basis of temperature variations.

If tektites formed on earth by meteorite and asteroid impact the variation in size might be explained by differences in explosive energy because of variation in size or speed of the impacting objects. Australites would have been the result of a very intense explosion with explosive energy enough to propel the molten material beyond the atmosphere, spreading it over an area the size of a continent. Other explosions would have been less severe, the molten material from none of them thrown high enough to be remelted as it re-entered the atmosphere. In some the explosive energy may have been insufficient to cause the molten material to pass beyond the atmosphere. However, if tektites are of impact origin, why have no mixtures of glass and partly fused materials been found?

Distribution and age

Any theory of origin of tektites to be valid must explain the distribution of tektites on the earth's surface in groups of widely different ages. Australites are found over two-thirds of the continent of Australia and may be present on an even larger area of the ocean bottom. They are the youngest known tektites and may have the widest distribution.

The tektites situated about the South China Sea, including the East Indies and the Philippines, extend at least as far west as Thailand. Similarities among these tektites suggest that they could all belong to one group; if they do their area of occurrence might exceed that of the australites. These tektites have been dated as middle Pleistocene.

The moldavites are believed to be weathering out of beds of Miocene age, and if this is true the area of their original distribution will probably never be known. The same is true for the bediasites, which are found for a distance of only 120 miles along the outcropping edge of the Jackson formation of Eocene age. The Ivory Coast tektites may possibly be older than the bediasites but this is not surely established. Little is known about their distribution.

The finding of tektites widely distributed in groups of widely different ages

with none forming between makes an earthly origin of tektites extremely unlikely unless tektites are actually the result of impact. Urey (1957) points out objections to tektites arriving as a swarm and suggests a different comet theory from that of H. E. Suess (1951). Whether or not Urey's objections to a tektite swarm are valid the writer does not know, but his treatment of the second period of melting (flow marks) seems inconclusive. If, however, the explosion produced by a comet was sufficient to drive molten material beyond the atmosphere where it solidified before again plunging into the atmosphere to be partly re-fused, then such an origin for tektites might have merit. The comet origin still does not explain the absence of glass with partly fused materials, and the same objection would apply to impact products from the moon.

Other Considerations

H. E. Suess (1951) found that the bubbles in tektites are very good vacuums and concluded that this is evidence that tektites either solidified in a near vacuum or that the bubbles were formed by water-vapour which was later absorbed by the glass. Friedman (1955) found the water content of tektites (0·002 to 0·008 per cent) to be very low in comparison to that of obsidian. During the tektite conference held in Washington D.C., 17–18 June 1957, Friedman displayed a melted tektite that vesiculated to two or three times its original volume. This tektite contained 20 p.p.m. of water.

D. Hubbard, U.S. National Bureau of Standards, also mentioned during the conference that it is not uncommon for shrinkage (vacuum) bubbles to form in glass during cooling. It seems likely, therefore, that the low pressure in bubbles is not pertinent in any theory of their origin.

The K^{40}–A^{40} age determination on tektites of Suess *et al.* (1951) and of Gerling and Yaschenko (1953) are inconclusive because they did not make determinations on the older tektites. Until the bediasites or Ivory Coast tektites have been investigated by this method, the K^{40}–A^{40} data so far obtained cannot be properly used in evaluating the origin of tektites. If it is found that argon-40 is present in these older tektites and that their ages are compatible with that of the strata from which they are derived, then evidence will be strong that the glass of the various tektite groups formed at different times. Such evidence would lend strength to a terrestrial origin and tend to discredit their formation by the destruction of a planet.

Results of studies of cosmic-ray induced radioactivities for Al^{26} and Be^{10} according to Ehmann (1957), indicate that tektites are of cosmic origin. However, only two samples of terrestrial material, one a pebble of granite from a conglomerate and the other an obsidian, have been examined, and until more samples of known geologic history, including material of sedimentary origin, have been analysed for comparison such a conclusion does not seem warranted.

Pinson *et al.* (1956) analysed one Philippine Island tektite by mass spectrometry for K, Rb and Sr, and concluded that it has a minimum age of 750×10^6 years, and therefore could not be of terrestrial age because "volcanic glasses (obsidians) older than Mesozoic are rare or unknown." They also stated "under terrestrial conditions a glass of tektite composition would be expected to devitrify in so great a time."

Devitrification of volcanic glass appears to be a continuous process which proceeds at a fairly uniform rate. MARSHALL (1954) indicated that diffusion of water into volcanic glass appears to be the best explanation for devitrification. He found a film of devitrification to be 7 μ thick in an Eocene perlite and only 1 μ thick in a Pliocene perlite. Most obsidian of Eocene age shows some evidence of devitrification, whereas Eocene tektites do not show the slightest sign of devitrification—this fact alone should be sufficient to show that comparing tektite glass with obsidian is unwarranted. Tektites were molten only momentarily, and during this short time it is unlikely that appreciable compositional change took place. The analyses by PINSON *et al.* therefore represent the material from which the tektite was fused, and if, as is strongly indicated, tektites are fused sedimentary rocks, this age determination is meaningless because of changes in chemical composition that took place during the weathering–sedimentation–lithification cycle.

SUMMARY

The tektite characteristics discussed in this paper versus the probability of certain origins are tabulated as follows, and it is hoped without bias:

	Terrestrial origin						Extra-terrestrial origin			
			Impact				Moon			
	Volcanic	Lightning	Meteorite	Asteroid	Comet (UREY, 1957)	Atomic bomb	Volcanic	Impact	Asteroids colliding with each other	Destruction of planet similar to earth
Two periods of melting	No	No	No	?	?	No	No?	Yes	Yes	Yes
Lechatelierite particles	No	Yes	Yes	Yes	Yes	Yes	No	No	No	Yes
Flow structure	No	?	Yes	Yes	Yes	Yes	No	Yes	Yes	Yes
Chemical composition	No	Yes	No?	No?	Yes	Yes	No	No	No	Yes
Shape	?	No	Yes	Yes	Yes	No	?	Yes	Yes	Yes
Size	Yes	No	Yes	Yes	Yes	No	Yes	Yes	Yes ?	Yes
Distribution	No	No	No	?	?	No				
Age	No	No	Yes	Yes	Yes	No	Yes	Yes	Yes	Yes

The reason for the wide distribution of tektites in individual showers mostly is the field of the mathematician and the astronomer, and not that of the geologist; therefore blanks are left in the above tabulation for distribution under extra-terrestrial origin. From this tabulation it is clear that a volcanic origin either from the earth or the moon needs no further consideration; likewise an origin from lightning or as impactites either from the moon or from collision of asteroids with each other has little support. An origin from the explosion on earth of a nuclear device is clearly not the explanation, even though such glass has characteristics of a tektite glass. The question arises: could an explosion on another planet have destroyed the planet and produced tektites?

Of the two remaining origins, *as determined from the examination of tektites alone* (and disregarding for the present their distribution), their formation by the destruction of a planet similar to the earth appears most likely. The impact theory does not explain two periods of melting unless molten rock was thrown beyond the earth's atmosphere and was remelted upon its return. The lack of tektites with partly fused minerals is against such an origin. However, it is possible these materials have been overlooked. Partly fused rock should remain fairly near to the point of impact because it would not have received the full force of the explosion. If the various points of impact are now far from land or covered by younger rock, such material could be present and not have been found.

Any explosive force or impact sufficient to disrupt a planet the size of the earth should form great spurts of gasified and completely liquefied rock, and these possibly formed comets by means of which tektites may have reached the earth. Even though this type of origin may appear attractive to geologists and geochemists, mathematicians and astronomers may be able to prove that such an origin is impossible. The writer is convinced that most tektites were originally sedimentary rock, that a terrestrial origin is most probable, but that the possibility of their formation from destruction of a planet similar to the earth should not be overlooked.

Obviously much work is needed, not only on tektites but also on rocks of the earth having similar composition, before the problem can be solved. Several samples of each of all the tektite groups should be investigated by each of the geochemical and other methods. Additional investigations are needed in all tektite localities to be sure that all groups are correctly dated, to see if partly fused materials have been overlooked, and to see if there are directions of change in composition, density, or size that may be significant. Perhaps if this is done, the origin of tektites may be solved.

Appendix

Publications on naturally occurring glasses:

Americanites, Columbia (Lleras Codazzi, 1915, 1925; Stutzer, 1926); Peru (Linck, 1926). Australites, Australia and Tasmania (Walcott, 1898; Fenner, 1934, 1935, 1938, 1940, 1949; Baker and Forster, 1943; Baker, 1944, 1946, 1955, 1956; Summers, 1913). Bediasites, Texas, U.S. of America (Barnes, 1940, 1956). Billitonites, Billiton (van Dijk, 1879; ver Beek, 1897; Easton, 1921); Borneo (Mueller, 1915); Bunguran, Natuna Archipelago (Krause, 1898); Java (Koenigswald von, 1935, 1936). Darwin glass, Tasmania (Dunn, 1916; David, Summers and Ampt, 1927). Empire, Georgia, U.S. of America (Barnes, 1956). Indochinites, Indo-China and adjacent areas (Lacroix, 1929, 1932, 1935). Ivory Coast tektites (Lacroix, 1934, 1935). Libyan Desert glass, Egypt (Caylton and Spencer, 1934; Spencer, 1939). Moldavites, Bohemia and Moravia, Czechoslovakia (Mayer, 1788; F. E. Suess, 1900, 1909, 1914). Philippinites including rizalites, Philippine Islands (Selga, 1930; Beyer, 1955). Familiarity with these publications as well as with most of the papers in the list of references is desirable before any theory of origin for tektites is proposed.

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The absorption spectra of tektites and other natural glasses

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Abstract—The absorption spectra of individual tektites known as australite, bediasite, indochinite, javanite, moldavite, and philippinite are compared in the 3000–26,000 Å region with that of Aouelloul glass, Libyan Desert glass, americanites, obsidian, perlite and an industrial welding goggle glass. The most characteristic feature of the absorption spectra of all these glasses in the range studied is the ferrous band with maximum in the 1·1–1·2 μ wavelength region of the near-infra-red. The spectra of these materials were not affected by X-ray treatment with the exception of an obsidian which showed an increase in the ultra-violet cut-off and growth in the ferrous band. The refractive indices (N_D) are compared and appear to be related to total iron content of the glass.

Introduction

The only prior investigations of the spectra of tektites were made by Stair (1955) and Houziaux (1956). Stair published transmittance curves for australites, bediasites, moldavites, philippinites and Libyan Desert glass in the ultra-violet, visible and infra-red regions while Houziaux measured the infra-red spectra of five tektites and two obsidians in the 2–24 μ range.

The spectra reported here agree well with those of Stair for tektites of the same type. However, there are some differences in interpretation of the data.

Experimental Details

The specimens were prepared by wafering with metal bonded diamond wheels or (in the case of smaller specimens) grinding down to the desired thickness with abrasives, and then polishing with cerium or tin dioxide polishing compound. The specimen thicknesses were measured with a Brown and Sharpe metric measure micrometer to $\pm 0{\cdot}001$ mm. All absorption spectral data were taken on a Cary Model 14M Recording Spectrophotometer Serial 66, with the exception of the obsidian data. The latter was obtained with a Beckman DU Spectrophotometer equipped with photo-multiplier attachment and with a filter to remove the ultra-violet back reflection in the 1000–1500 mμ region. The X-ray irradiations were made using a Picker Industrial X-ray unit designed for continuous operation at 50 pkV, 40 mA. The tube used was a Machlett AEG50T with tungsten target and beryllium window. The irradiations were at 45 pkV and 35 mA, 6 cm from the X-ray tube window. The dosage rate is in the order of 2–3 $\times 10^6$ r/hr in air at the surface of the wafer. The X-ray irradiations were carried out at room temperature as were all spectral measurements. All samples were wrapped in aluminium foil during irradiation and transferred to the spectrophotometer in a darkened room in order that any possible X-ray induced effects would not be bleached if sensitive to light. The refractive index measurements were made using a Bausch and Lomb Precision Refractometer and a sodium vapour lamp as the light source.

Results and Discussion

A. Natural glasses

A uniformly coloured optical-quality obsidian collected in the Arroyo Hondo, Valles Mountains region of New Mexico by R. L. Smith of the U.S. Geological Survey was spectrophotometered as shown in Fig. 1. The spectrum consists of a

* Pittsburgh Plate Glass Company Fellowship

sharp cut-off in the ultra-violet, a weak, broad band of unknown origin in the 500–700 mμ region of the visible spectrum and a very broad band with a maximum at 1200 mμ due to ferrous iron (Weyl, 1951). Upon X-irradiation there was a rise in the ferrous band suggesting that some of the ferric iron present was reduced by this treatment. The growth of the ferrous band is indicated in the small inset in Fig. 1. The peak underwent no further increase after 17 hr of irradiation. The ultra-violet cut-off moved to longer wavelengths upon irradiation indicating the growth of bands at a shorter wavelength. The slight absorption increase in the

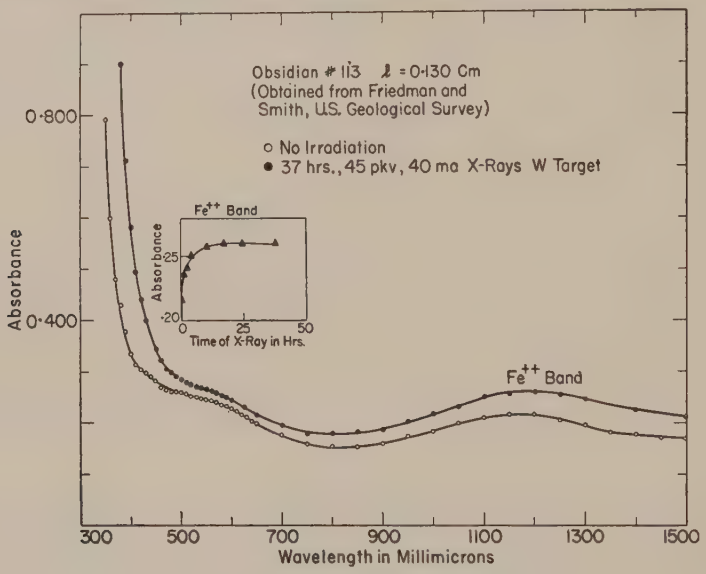

Fig. 1. Spectrum of an obsidian from Arroyo Hondo, Valles Mountains, New Mexico, before and after X-ray treatment.

500–700 mμ region seems mainly due to overlap of the short wavelength band(s). The growth of the ferrous band and its saturation upon X-ray treatment is not understood as yet. It was thought that tektites might exhibit growth of the ferrous band upon treatment with ionizing radiation but this was not found to be the case as will be discussed later.

An investigation of the Arroyo Hondo obsidian in the region 15,000–26,500 Å indicated that if any hydroxyl groups were present they were below the optical detection limit in the spectral region under study.

The upper spectrum in Fig. 2 is for a specimen of perlite, a hydrated obsidian which contains over 4 per cent water (Friedman, 1957). This specimen was collected by R. L. Smith from Bear Head, Jemez Mts., New Mexico. Ross and Smith (1955) have discussed the water content of similar perlites in detail. The spectrum is characterized by the broad ferrous band with maximum at 1·22 μ and by three vibrational bands, two of which are related to the hydroxyl group (Herzberg, 1945). The band at 1·41 μ is probably due to the combination of the ν_1 and ν_3 fundamental OH stretching vibrations. The sharp band at 1·91 μ is probably due to the combination of a stretching and bending vibration (ν_2 and ν_3).

A band at 2·22 mμ (also seen in the Arroyo Hondo obsidian where hydroxyl was not detected) could be the first overtone of the Si—O stretching vibration. However, it does not appear in all the spectra studied here or in all silica that has been investigated so its origin is still in doubt.

The lower spectrum in Fig. 2 is that of an americanite from Peru obtained from FRIEDMAN who found it contained 0·23 per cent water. The bands due to the hydroxyl group are hardly noticeable in the drawing although present. The 2·22 μ band however is still intense, indicating that it is unlikely that this band is related

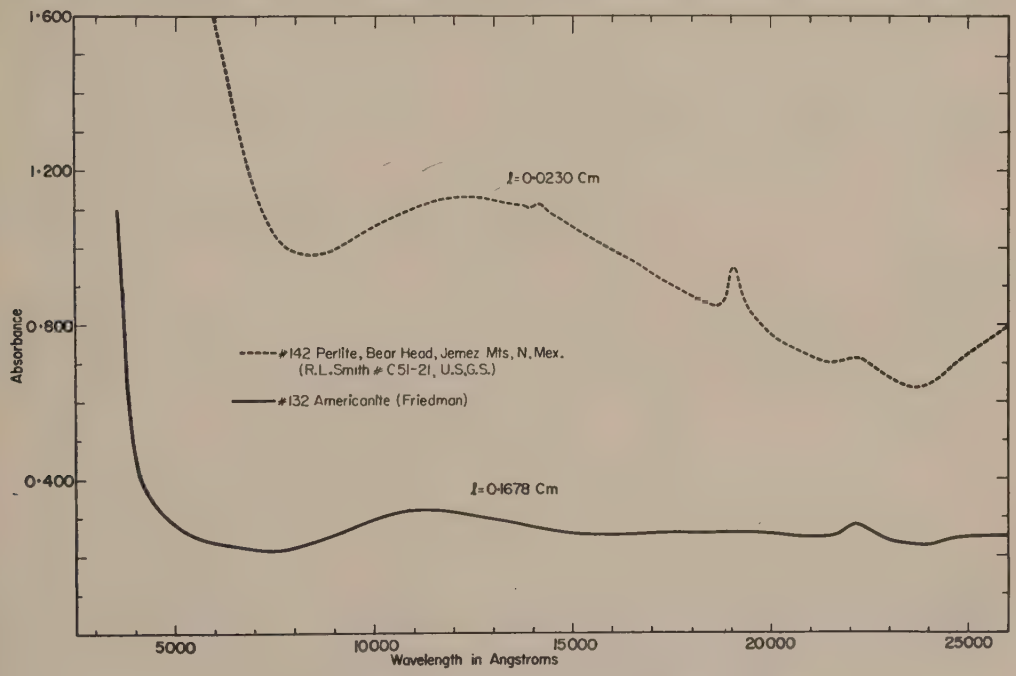

Fig. 2. Comparison of the spectra of two volcanic glasses.

to the hydroxyl group. The ferrous band is this material is also low. This is in agreement with LINCK (1926) who found the ferrous plus manganous oxide content to be 0·96 per cent (see Table 1). LINCK believed the material to be a tektite. As brought out by several workers present at the Tektite Conference, americanites are now known to be a rare type of obsidian. Two other americanites were investigated. Both specimens were gifts of the Rijksmuseum von Geologie en Mineralogie of Leiden and were from Macusani, Peru. They both contained many diverse mineral inclusions as described by LINCK. One was pale tan and transparent in thin section (specimens no. 173 and no. 176). The other (specimen no. 182) contained red opaque inclusions. These inclusions are barely resolved at a magnification of 160 and are directionally streaked out in the glass. The spectrum of the pale coloured americanite is illustrated in Fig. 3. The absorption coefficient (k) of the ferrous band is only slightly higher for this specimen (no. 176 and a thinner section no. 173) than for the material obtained from FRIEDMAN as shown in Table 1. The thicker specimen shows a small 1·41 μ band indicating that hydroyyl ion is

present as well as the band at 2·22 μ. These two bands are too weak to be seen in the thin section. The non-uniformity of this material is brought out by the difference in the k for the ferrous band in the two samples from the same specimen.

The spectrum of Aouelloul glass from Adrar, Western Sahara (specimen no. 180) is illustrated in Fig. 3. This material was collected by Cassidy and is most likely an impactite produced by the heat developed in terrestrial material by meteorite collision. Since Cassidy was unable to find any meteoritic material even with the aid of mine detection equipment, the type of meteorite that produced the

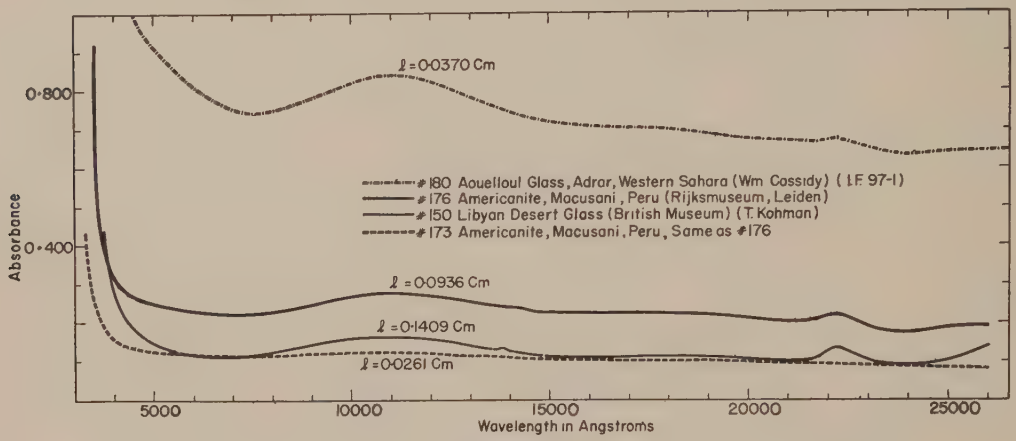

Fig. 3. Comparison of the spectra of Aouelloul glass, of Libyan Desert glass, and of an americanite.

crater is unknown. This fact makes it important to compare the trace impurity content of the glass to that of surrounding silicate material. In this regard Smith and Hey have analysed two sandstone samples from the crater. They found that Aouelloul glass contains more aluminium and potassium than the sandstones in the crater. Iron and stony meteorites are low but tektites are high in these two elements. In view of this and the nature of the glass which has colourless areas of silica-rich material and incorporates incompletely melted grains (furnished by the sand or sandstone) these authors then suggest that the crater is a tektite crater and the Aouelloul glass is an impactite made from mixing of molten tektite with local sandstone, the composition being intermediate between a moldavite and the local sandstone. Examination of thin sections of Aouelloul glass in this laboratory confirms the earlier observations of these authors that small streaky brown patches, birefringent crystal fragments and many spherical cavities of diverse size are present. No nickel–iron inclusions were observed in the glass.

Friedman found 0·02 per cent water present in another portion of the same specimen (his number 97-1). However, with the 0·0370 cm thick specimen, this amount of hydroxyl was not optically detectable in the region under study. A good refractive index measurement could not be obtained due to the inhomogeneous nature of the material.

The spectrum of Libyan Desert glass indicates that the ferrous content is quite low (Fig. 3). This is in agreement with analysis (see Table 1). That hydroxyl is

present is quite evident from the spectrum. FRIEDMAN finds a minimum of 0·064 per cent water in a specimen of this glass. This is an order of ten higher than that found in any of the tektites and three times as high as the water content of Aouelloul glass. The refractive index found for the specimen studied (1·46056) is slightly lower than $N_D = 1{\cdot}4624$ reported by CLAYTON and SPENCER (1934).

B. Tektites

The spectrum of a moldavite (no. 74) as shown in Fig. 4 is characterized by the ferrous band and by a broad band of unknown origin in the 1·6–2·2 μ region.

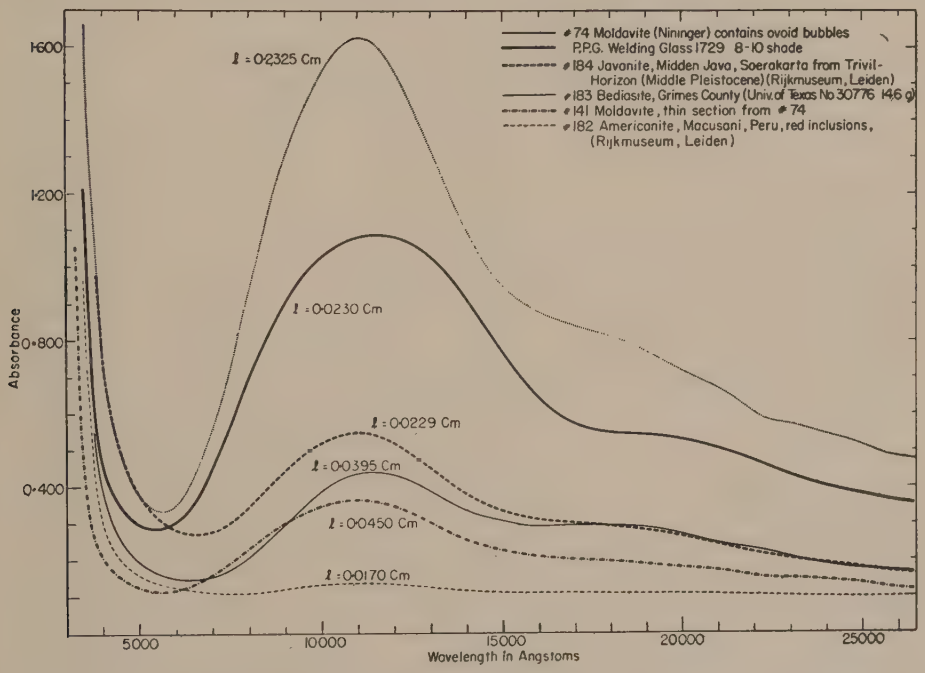

Fig. 4. The absorption spectra of three tektites, an americanite, and an artificial iron-containing glass.

Comparison with a thin section of the same specimen (no. 141) suggests that the material does not obey Lambert's law. However, the spectra were not corrected for reflection losses, so that the difference in absorption coefficients (Table 1) may result from this error.

STAIR published the spectrum of a tektite from Empire, Georgia. A wafer was obtained from one of two specimens which were purchased by the Smithsonian Institution (tektite no. 1396). Both these tektites are flattened ellipsoids similar in form and colour to some moldavites. The spectrum is not shown here but it is very similar to that of the moldavite with the ferrous band maximum at the same wavelength and the broad band at 1·6–2·2 μ present. The absorption coefficient of the ferrous band maxima was 8·40 cm^{-1} which is nearer that of the moldavite than any of the other tektites studied (see Table 1). The "Georgia" specimen examined in thin section had inhomogeneity similar to that of a moldavite

and similarly contained lechatelierite inclusions. One is inclined to regard the Empire, Georgia tektites as moldavites until concrete evidence of their locality and the stratum in which they are found is forthcoming.

The spectra of a javanite, bediasite and an americanite are also included in Fig. 4. The latter two tektites have spectra very similar to that of the moldavite; the major differences between these tektites is only one of intensity of the bands as shown by the k's of the ferrous band in Table 1. The australite, indochinite,

Table 1. Absorption coefficients of Fe^{++} band maxima, Fe contents and refractive indices of tektites and other natural glasses

Sample no.	Glass	Fe^{++} band 1·11–1·20 μ (k cm^{-1})	Chemical analyses (wt. %) FeO	Fe_2O_3	N_D
150	Libyan desert	1·15	0·23*	0·11*	1·46056*
113	Obsidian	1·66	—	—	—
113	Obsidian + 37 hr X-ray	2·00	—	—	1·4863**
132	Americanite	1·91	0·96†	—	1·48552§
176	Americanite	2·94	0·96†	—	1·48552**
173	Americanite (same as 176)	4·60	0·96†		1·48552**
74	Moldavite	7·01	1·13–3·36*	0·14–2·05*	—‡‡
141	Moldavite (thin section of 74)	8·11	1·13–3·36*	0·14–2·05*	—‡‡
182	Americanite (red inclusions)	9·41	0·96†		1·48584††
183	Bediasite	11·11	3·36–4·64*	0·37–0·45*	1·49065††
134	Australite	18·33	3·11–5·30*	0·32–0·85*	1·50855§
133	Philippinite	21·79	3·03–5·32	0·59–2·03	—‡‡
180	Aouelloul glass	22·70	0·05–1·72‡	0·45–1·45‡	—‡‡
184	Javanite	23·93	4·46*	0·83*	—‡‡
136	Indochinite	28·00	3·59–5·63*	0·06–0·37*	—‡‡
—	P. P. G. Welding no. 1729	47·39	—	—	
142	Perlite (C51-21)	49·17	—	—	1·49936**

* Barnes (1940).
† Linck (1926) MnO + FeO = 0·96.
‡ Smith and Hey (1952).
§ at 27·2°C.
** at 27·6°C.
†† at 26·5°C.
‡‡ index of refraction not uniform enough for good reading.

and philippinite spectra in Fig. 5 are similar to the spectra of the other tektites in Fig. 4.

The tektite spectra resemble those of iron-containing commercial glasses. The welding goggle glass in Fig. 4 differs in having a higher ferrous content than any of the tektites. The tektites differ from obsidian, americanites, and Libyan Desert glass in showing no hydroxyl bands in the region under study. This region does not, of course, cover the fundamental bands. However, this work gives a rough indication of hydroxyl content in the non-tektite samples and in agreement with Friedman shows that with these specimens, $>0{\cdot}06$ per cent water is detectable.

In Table 1 the glasses are tabulated in order of increasing absorption coefficients

of the ferrous band. It is seen that these coefficients agree in general with the range of FeO analyses of other investigators. Only the lowest and highest values of several analyses are given rather than averages. The Aouelloul glass gives the only poor agreement and this can be attributed to the irregular nature of the colour and the presence of opaque regions which raise the absorbance.

Indices of refraction are also listed in Table 1, and in general increase with total iron content. It was impossible to obtain indices for the moldavite, the so-called Empire, Georgia tektite, the philippinite, Aouelloul glass, the javanite

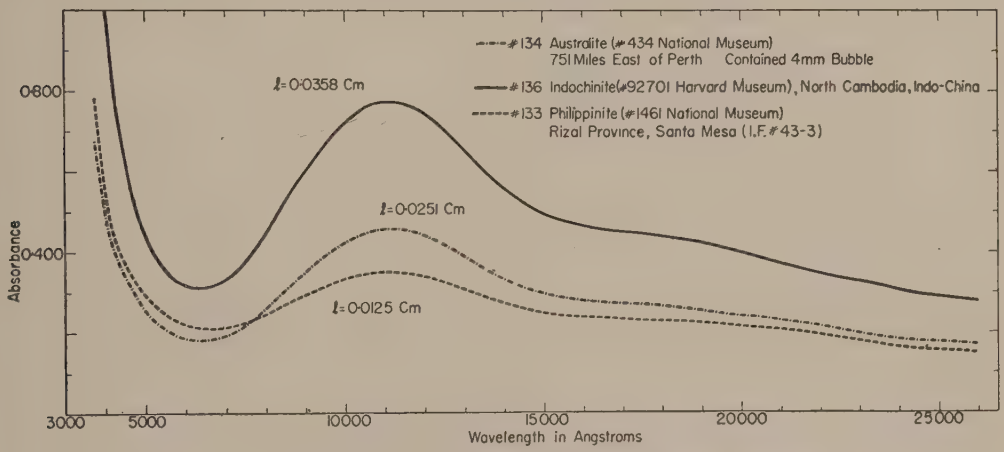

Fig. 5. The spectra of three tektites from the eastern hemisphere.

and the indochinite because of the inhomogeneous character of these glasses. The indices of these glasses have been determined earlier by usually less precise methods and are tabulated by Barnes (1940). The average value listed for twenty indochinites is 1·5072, with a range of 1·4986 to 1·5148, for thirty-eight moldavites is 1·4888, and for one philippinite is 1·5130. These are in general agreement with the iron contents and with the ferrous content. One might conclude that the ferrous content is an indication of the total iron content and would thus correlate with refractive index. Upon examination of ten welding glasses of differing iron content this was found to be the case in nine instances.

Stair suggests that the "relatively high infra-red transmittances of the tektites in the spectral region of 1000 to 2000 millimicrons indicates that much of the iron may be in the unreduced state (Fe_2O_3) or in some equivalent combination as regards infra-red spectral absorption." The transmittances in the 1200 mμ region related to the presence of ferrous ion in the glass are actually relatively low as shown in Fig. 7 of Stair's paper and as shown here (as high absorbance). The optical investigation thus substantiates the earlier chemical analyses that tektites are high in ferrous iron as shown by the rough agreement between the intensity of the ferrous band and the analyses.

C. Irradiation of natural glasses and tektites

X-ray treatment of the australite, indochinite, philippinite, javanite, bediasite, Libyan Desert glass, Aouelloul glass, americanite (no. 173), and the perlite gave

no change of intensity in the region of any absorption bands. These irradiation times were sufficient to colour the usual silicate glasses deeply. There was some change in the moldavite spectrum; however, this can be accounted for by the rise in the ultra-violet region and thus the slight increase in the ferrous band region is probably not due to increase in the ferrous iron content.

Heating of a moldavite to near its melting point did not change the spectrum. This was also found to be the case for the welding glass. One can then conclude that the iron content of these glasses offers radiation protection similar to that given commercially by cerium and that cosmic-ray and ultra-violet radiations are without effect on these glasses. Changes in the ferrous spectrum upon X-irradiation appear also too small to detect optically.

One concludes that thermoluminescence studies will offer more promise than spectral methods for investigations of possible changes in the tektites from high-energy radiation, whether it be from cosmic rays, ultra-violet rays, or from natural radioactivities contained in the material. F. G. Houtermans has reported some preliminary results on thermoluminescence in tektites at the Tektite Conference.

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Mr. Herbert L. Smith ran a major portion of the absorption spectra curves and took the measurements of indices of refraction.

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Impact effects and tektites*†

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Abstract—Data on the composition, form and distribution of tektites are examined in the light of our knowledge of high speed impact phenomena to see how strong the evidence is for an impact origin. It is concluded that tektites could be of impact origin, but the arguments are inconclusive. A strong argument against such an origin, is the lack of nickel–ferrous materials within the tektite.

Are tektites fusion products formed at the time of impact when large meteorites strike the earth? Their composition, shape and distribution suggest that they could have an impact origin, but the evidence is not really critical or definitive. A strong argument against such an origin is the lack of nickel–iron materials within the tektite, but this argument, too, is not incontrovertible. In this paper we critically examine data on the composition, form and distribution of tektites in the light of our knowledge of high speed impact phenomena to see just how strong the evidence is for an impact origin.

If tektites are to be explained as detritus of earthly impacts of meteorites we must first determine whether molten material can, in fact, be splashed from the crater formed by the contact. Second, the composition of this fused material must be that of the rock and soil indigenous to the area, and should very probably contain meteoritic material as well. Third, the individual pieces of detritus could reasonably be expected to assume shapes characteristic of rapidly cooled, severely agitated molten material. And, finally, their distribution on the ground within a particular strewnfield should resemble the distribution which might arise from an impact.

Could a meteorite strike with sufficient velocity to fuse the earth about its point of impact? Yes. Meteorites enter the atmosphere with velocities that range from 12 km/sec to 70 km/sec. The average velocity is about 17 km/sec (Whipple and Hughes, 1955). Small meteorites are rapidly decelerated by the atmosphere so that they strike at rather low velocities, about 1000 ft/sec at most, and at such velocities there will be no vitrification of the sand and rocks. On the other hand, large meteorites greater than 1000 tons will not be appreciably decelerated and will strike the earth with such great velocities that intense heat is generated, the sand and rocks are melted, and much molten material thrown out. We have not yet been able to fire projectiles at such velocities in the laboratory so that we do not know for certain that this is what takes place. But theoretical calculations by Gilvarry and Hill (1956) indicate that the temperatures reached by the target would be sufficiently high for liquid material to be formed. Our best clue as to what can happen is the thermally altered sandstone found around the Arizona Meteorite Crater and the Wabar Craters in Arabia (Nininger, 1956; Spencer, 1933). This

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cindery and porous material, impactite, contains inclusions of nickel–iron. Since an impacting meteorite must lose much of its mass to the surrounding soil, this is to be expected. Tektites are solid and vitreous. The composition of tektites has been discussed in great detail by Fenner (1934, 1935, 1938, 1940, 1949), Barnes (1940), and others. The composition of any tektite yet analysed may be duplicated reasonably well by some particular sedimentary rock;* thus, tektites could be metamorphosed country rock. The individual compositions of members of a particular tektite swarm are similar, although there are differences in composition between swarms. This fact indicates that each swarm has a similar origin, but does not argue against the theory of impact origin. It must be noted that an igneous glass of composition similar to that of tektites has not been found.

Tektites do not contain significant amounts of iron and nickel; in particular, they have no inclusions of nickel–iron bits. While some compositional features support an impact hypothesis, the lack of nickel–iron inclusions to some degree opposes it. The lack of such inclusions suggests that if tektites came from an impact crater, they must be assumed to represent material that did not contact the meteorite that bored into earth; or, the meteorite that fell was essentially free from metallic inclusions. Large amounts of intimately mixed material would be thrown out and it is surprising that we do not find some of it in the strewnfields of tektites. Impactite is quite common near the Arizona meteorite crater, the Henbury craters in Australia, and the Wabar craters in Arabia. Near these craters we never find tektite-like, nickel-free fused silica blobs.

The shapes of tektites have been thoroughly described by Fenner (1934, 1935, 1938, 1940, 1949) and Barnes (1940) and their descriptions need not be repeated here. The shapes are entirely consistent with an impact origin. Fenner (1934) observed in the smoke of coal-burning railway engines, blobs or bombs of vitreous material, presumably silicates; and from a critical study of these smoke bombs he concluded that the primary shapes are those that are generated when glassy materials solidify while being blown about: spheres, boats, canoes, dumbbells, buttons.

Worthington (1908), perhaps, throws the most light on what we may expect when splashes from liquid impacts break up into blobs or drops. From his discussion it is apparent that shapes resembling boats, canoes, dumbbells, etc. could all reasonably be formed. As the liquid or molten rock or sand is spewed forth from the impact crater it is first in the form of a liquid sheet, much as a wave is before it breaks. The sheet, as it moves out from the crater, begins to collapse into columns and droplets. The collapse of such columns was treated theoretically many years ago by Plateau (Worthington, 1908), who showed that a cylindrical column of liquid would collapse into a group of droplets none of whose diameter was larger than the diameter of the column. It is easy to see how during collapse many forms of tektites arise, particularly if the material within the column is fairly viscous. The dumbbell-shaped tektites are simply the segments of an incompletely collapsed column. Teardrop shapes are formed in much the same way.

* Evidence presented at this conference on the presence of trace elements suggests that their abundances may be incompatible with a terrestrial origin but the data are not definitive.

Many tektites also appear to have undergone a second shaping (Fenner, 1935). Flanges circumscribe the teardrops, and the buttons have well developed rims and appear to be remnants of what were once perfect spheres. These features must have developed during flight, probably high velocity flight: velocities so high, several kilometres per second, that ablation would take place. During impact, material is ejected from the crater with a velocity great enough for the observed ablation to take place. In fact, the velocity of the ejecta is sometimes greater than that of the impacting missile. It is unlikely that secondary shaping required more than a few minutes. The secondary shaping, ablation, and formation of flanges and rims is also consistent with an impact origin.

The final point we must consider is the distribution of tektites. Here we have two features: the global distribution and the distribution within a strewnfield. Viewed globally there are some ten large areas where tektites are found. These are fairly uniformly distributed over the surface of the earth, as are the sites of impact of large meteorites. This random global distribution is not inconsistent with an impact origin.

The distribution within a particular strewnfield is best known for the australites (Fenner, 1935). They cover a large roughly elliptical area about 2000 miles long and 1500 miles wide. The bediasites, in Texas, are found in an area about 10 miles long and 5 miles wide (Barnes, 1940). No crater is associated with any known field. If the impact were vertical, then one would expect the debris to be distributed concentrically around the point of impact. Certainly no such juxtaposition of crater and tektites is found in semi-arid regions where exceedingly long periods of time would be needed to erase the evidence of a crater. Meteorites of course could also strike obliquely. Under these circumstances large amounts of thermally altered country rock or sand would be thrown forward and upward with great velocity and become scattered over a large area. If the angle were very oblique the meteorite would scour a shallow trough which might easily become obliterated in time. The meteorite itself would ricochet and might be carried way beyond the principal field of ejecta or it might fall short of the field. An impact origin for tektites does not necessarily imply that strewnfield and crater will be found side by side, for the impact might well, and probably would be, an oblique one.

Now how does the evidence add up? On the basis of composition, shape, and distribution, tektites could be of impact origin, but the arguments are inconclusive. A strong argument against an impact origin lies in the lack of nickel–ferrous materials within the tektite. Perhaps, however, the detritus was differentially distributed so that the fused rock in which meteoritic material was interspersed may have splashed to an area apart from that in which the tektites landed. Tektites would then be material untouched by the meteorite although heavily affected by the heat and pressure of the impact. Such a division of material does not seem unreasonable. More data on tektites may help to resolve the dilemma but the avenues of approach are not apparent.

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Dynamical limits on a lunar origin for tektites*

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Abstract—Theories regarding the origin of tektites are reviewed and it is proposed that the moon is the most likely source for these objects. Detailed calculations show that Whipple's model for the ejection of tektites from the moon makes it possible to reproduce their observed distribution over the earth's surface.

1. Introduction

Since tektites were first discovered, and their shapes and compositions analysed, several theories have been formulated to account for their origin. These theories can be classified into two groups, one assigning to the tektites a terrestrial, the other an extra-terrestrial origin.

Because of the peculiar physical and chemical properties exhibited by tektites, the theories assigning them a terrestrial origin must postulate some sort of catastrophic event. For example, Spencer (1933) suggested that tektites may be debris resulting from the fall of a meteorite on the earth's surface; Barnes (1940) proposed that tektites could be formed when lightning strikes sedimentary rocks, a theory which he later discarded in favour of an extra-terrestrial source (Barnes, 1951); Urey (1957) believes that they may be formed when a comet strikes the earth. Among the theories proposing an extra-terrestrial source is Nininger's (1947) suggestion that tektites may be "chips from the moon," ejected from the lunar surface by meteoritic impacts. Later Suess (1951) proposed that these objects may be fragments of the head of a comet that disintegrated in the vicinity of the earth. Several authors consider that tektites are a special type of meteorite that could have originated at the disruption of a planet similar to the earth (see, for example, Stair, 1956, and Cassidy, 1956).

The list of theories presented above is by no means exhaustive, but is sufficient to give an idea of the variety of origins that have been suggested. Recent research permits us to discard most of these theories, and this paper proposes one that can account satisfactorily for most observed facts about tektites.

A terrestrial origin of tektites does not seem compatible with their chemical composition. The work of Ehmann (1957) on the radioactive isotopes Al^{26} and Be^{10} gives evidence that tektites come from an extra-terrestrial source. Friedmann's (1958) measurements of the water content of several types of glasses show that no process imaginable could produce, on the earth, a glass as anhydrous as tektites. The constancy of their content of potassium and rubidium found by Pinson and Herzog (1958) is also inconsistent with a terrestrial origin. Finally, the physical appearance of tektites indicates that they have undergone a second heating,

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possibly because of their fast passage through the earth's atmosphere, which is consistent with an extra-terrestrial origin. Urey's (1957) comet-collision theory suggests an ingenious way to account for the appearance of tektites. It proposes that the flow marks were produced by a blast of hot gases passing over the slowly moving tektites.

When we consider those theories that assign to the tektites an extra-terrestrial origin, we find that their observed distribution over the earth's surface greatly limits the possibilities. Tektites are found in relatively localized areas, a well studied example being that of Australia (Fig. 1). This localized distribution

Fig. 1. Distribution of australites (according to Fenner). Shaded areas indicate the regions where australites were found. None were found north of the line AA'.

suggests that tektites reached the earth in a rather compact cloud. The same distribution would obtain if tektites arrived in the form of large aggregates which break up upon entering the earth's atmosphere. However, the physical appearance of the tektites would be difficult to explain on this basis. Since such a cloud is held together only by gravitational forces, and since these forces would be very small—we are dealing with bodies whose masses, on the average, are of the order of from 10 to 100 g—it follows that the cloud would retain its compactness for only a very short period of time, of the order of a few hours. Unless all particles in the cloud travel with the same velocity, a very unrealistic assumption, a velocity range of a few centimetres per second would be sufficient to disrupt the cloud in a matter of days. In other words, to reproduce the observed distribution of tektites on the earth's surface, such a cloud would have to encounter the earth very shortly after it began its travel through space; otherwise, it would have expanded enough to cover uniformly an entire hemisphere. This argument gives very strong evidence against the possibility that tektites may have originated from the disruption of a planet or a comet head.

One theory that has so far escaped criticism is that the moon is the source of tektites. A lunar origin is not, to my knowledge, inconsistent with any of the physical and chemical properties of tektites so far studied. We still need to prove, however, that the ejection of tektites from the moon is compatible with their observed distribution on the earth.

2. Physical Model of the Ejection Process

The calculations in this paper stemmed from a suggestion made by F. L. Whipple regarding the probable circumstances whereby lunar impactites, if such exist, might be produced by meteoritic collisions on the surface of the moon. The basic idea concerns the nature of the ejection of material from the lunar surface as a function of the angle of ejection from the normal to the surface. A high velocity body forming a crater would produce explosive gaseous ejection normal to the lunar surface. At extremely low angles to the surface, as suggested by Gold's (1955) qualitative study of the phenomenon, the ejection procedure would be a horizontal pushing and general crushing of material. At intermediate

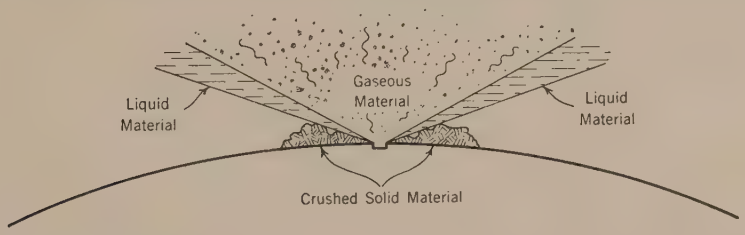

Fig. 2. The Whipple–Rinehart model for the ejection of impactites from the lunar surface.

angles the nature of the ejection would change systematically, from high temperature gaseous explosions normal to the surface to larger and larger particle ejection and lower and lower velocities, until the pushing phenomenon parallel to the surface would predominate. Impactites in which lunar material had become liquefied by the explosion, but was still being ejected at moderate velocities so that droplets might cool in space, would then be produced in a conical ring of fairly small angular thickness (see Fig. 2). Under these circumstances, the potential tektites produced by the crater-forming collision would be ejected in a cone of fairly narrow angular thickness.

The present paper, therefore, concerns the possibility of encounter of such a conical surface of tektites with the earth on the assumption that the velocity range of the material in the cone is appreciable. Specifically, we computed the shape and size of the regions of fall of lunar impactites on the earth's surface, and the distribution of the falling material within them.

3. Mathematical Formulation

These calculations were carried out on a high speed digital computer by means of which 6 orbits/min could be computed with an accuracy better than 0·1 per cent. The velocities of ejection adopted ranged between the velocity of escape from the moon (2·38 km/sec) and 5 km/sec, with intervals of 0·05 km/sec to 0·4 km/sec. All the material was assumed to be ejected in a 5° cone, whose sides made angles of 15° and 20° with the horizontal.

The problem is, mathematically, very straightforward. I adopted a geocentric system of rotating coordinates, with the plane of the earth–moon orbit coincident with the x–y plane, and the moon always on the x–axis. To facilitate the calculations, the moon was assumed to move in a circular orbit, and the gravitational

field of the sun to remain constant during the time of travel of the tektite. Calculation of their paths by numerical integration of the equations of motion, took into account the gravitational attraction of the sun, earth, and moon, and the coriolis and centrifugal forces that arise when rotating coordinates are used.

Let F = total force per unit mass acting on the tektite
k = gravitational constant
M_E = mass of the earth
M_M = mass of the moon
r = distance to the origin of the coordinate system
ξ = earth–moon distance,
v = linear velocity of the tektite
w = angular velocity of the moon in its orbit
x, y, z = position coordinates.

Then

$$\vec{F} = \vec{G}_M + \vec{G}_E - 2(\vec{w} \times \vec{v}) - \vec{w} \times (\vec{w} \times \vec{r}), \tag{1}$$

where

$$\vec{G}_E = \frac{kM_E}{r^3}\vec{r}; \qquad \vec{G}_M = \frac{kM_M}{[(\xi - x)^2 + y^2 + z^2]^{3/2}}\vec{r} \tag{2}$$

After the force acting on the tektite at a point $\vec{P}$ is calculated, we use this value to compute the new position $\vec{P^1}$, after an arbitrary elapsed time Δt by means of the formula:

$$\vec{P^1} = \vec{P} + \vec{v}\,\Delta t + \vec{F}\,\frac{(\Delta t)^2}{2} \tag{3}$$

The velocity $\vec{v^1}$ of the tektite at point $\vec{P^1}$ is computed from the expression:

$$\vec{v^1} = \vec{v} + \vec{F}\,\Delta t \tag{4}$$

Next we use equation (1) again, with the values appropriate to point $\vec{P^1}$; we insert the new value of $\vec{F}$ in equations (3) and (4), and proceed by iteration until the tektite hits the earth or is definitively lost.

4. Computations and Results

Even though the problem is very simple mathematically, computationally it becomes rather formidable because of the large number of variables involved: the three position coordinates of the point of ejection, the three components of the initial velocity, the angle the ejected material makes with the horizontal, and the position of the sun relative to the earth–moon system. A detailed survey of all the parameters would result in about 20 hr of work on an IBM 704 computer. Circumstances required us to choose the initial conditions very carefully in order to reduce the computation time to a maximum of 3 hr. This gave a clear answer to the problem in question, although the results were a coarser-than-ideal survey. Further calculations would improve the preciseness of the answer but would not modify our general conclusions.

Fig. 3 shows an x–y projection of the points of ejection investigated, and Table 1 lists their coordinates. The invisible half of the moon was ignored because, although it is possible that tektites ejected from this region may reach the earth, they would take a long time in doing so, and they would be drawn far apart from each other during their travel in space. Of the visible half of the moon only the part above or below the orbit plane needed to be considered because of the symmetry of the situation.

Fig. 3. Projection on the x–y plane of the points of ejection investigated.

The first series of calculations investigated points 1 through 9 and 11 through 13 (Fig. 3). Table 2 shows the results which helped determine the choice of initial parameters for the second survey. It immediately became apparent that there was a region on the moon from which "hits" could be expected, and a third and fourth survey were carried out to delimit this region accurately.

To determine how the falling material would be distributed over the earth, several orbits were computed from each starting point, covering reasonable ranges of ejection velocities and angular distributions. These calculations show that some points, like 1 and 14, give fairly localized falls, while matter ejected from 11, for example, would cover uniformly more than half the earth.

Our calculations allow us to tell whether a certain point of ejection will give a localized fall or a widespread one. However, we do not have enough information to predict which type would predominate statistically.

Table 1. Points of ejection from the lunar surface

Point no.	x (10^5 km)	y (km)	z (km)
1	3·840	1679	0
2	3·832	1229	0
3	3·827	450	0
4	3·827	−450	0
5	3·832	−1229	0
6	3·840	−1679	0
7	3·833	1064	869
8	3·829	389·5	869
9	3·829	−389·5	869
10	3·833	−1064	869
11	3·838	614·5	1505
12	3·838	−614·5	1505
13	3·840	0	1700
14	3·838	1578	449·8
15	3·837	1364	869
16	3·838	1064	1229
17	3·839	711·8	1505
18	3·841	318·1	1679
19	3·840	1622	449·8
20	3·837	855·8	1331·3
21	3·840	224·9	1679
22	3·841	1712	0

We could say, very roughly, that both types of fall should be expected in equal numbers.

To determine the relative abundances of tektites at points inside a region of fall we need to make two assumptions: One regards the amount of material that is ejected from the lunar surface as a function of the velocity; the other is the amount of material ejected as a function of the angle of ejection. The necessity for such assumptions is rather unfortunate because there are no experimental data available on the subject, and any distribution adopted must necessarily be arbitrary. For the purposes of computation I chose a Gaussian distribution of velocities, with a maximum at $V = 2{\cdot}5$ km/sec and precision index $= 1{\cdot}5$, so that over two-thirds of the material is supposed to be ejected with velocities between 2·0 and 3·0 km/sec; however, other plausible distributions would not affect the general results.

In agreement with an analytical treatment of T. E. Sterne (1957), I found that particles leaving the sphere of influence of the moon with very low velocity ($v < 0{\cdot}2$ km/sec) do not strike the earth. At slightly higher velocities the probability of a hit increases rapidly, and begins to decrease when $v > 0{\cdot}7$ km/sec (corresponding to a velocity of ejection of 3 km/sec). In all cases it is also true

Table 2. Result of the first survey

Point of ejection	Initial velocity (km/sec)			Value of y when $x = 0$ (km)	Value of z when $x = 0$ (km)	Remarks
	V_x	V_y	V_z			
1	−2·275	0·0871	−1·256	1050	−80,500	
	−2·516	0·0224	−0·650	500	−27,000	
	−2·600	0·0	0·0			Strikes the earth
	−2·516	0·0224	0·650	1050	80,500	
	−2·275	0·0871	1·256	500	27,000	
2	−2·014	−1·062	−1·256	−70,751	−105,080	$x = 4555$*
	−2·191	−1·239	−0·650	−72,000	−49,000	
	−2·251	−1·300	0·0	−73,000	0	
	−2·191	−1·239	0·650	−72,000	49,000	
	−2·014	−1·062	1·256	−70,751	105,080	$x = 4555$*
3	−1·213	−1·927	−1·256	−174,056	−127,311	$x = 139,657$
	−1·278	−2·168	−0·650	−186,419	−65,198	$x = 116,553$
	−1·300	−2·251	0·0	−190,600	0	$x = 108,421$
	−1·278	−2·168	0·650	−186,419	65,198	$x = 116,553$
	−1·213	−1·927	1·256	−174,056	127,311	$x = 139,657$
4	−1·213	1·927	−1·256	221,680	−131,982	$x = 465,160$
	−1·278	2·168	−0·650	245,867	−68,228	$x = 474,752$
	−1·300	2·251	0·0	254,376	0	$x = 478,191$
	−1·278	2·168	0·650	245,867	68,228	$x = 474,752$
	−1·213	1·927	1·256	221,680	131,982	$x = 465,160$
5	−2·014	1·062	−1·256	304,359	−125,608	$x = 349,402$
	−2·191	1·239	−0·650	329,537	−64,900	$x = 244,111$
	−2·251	1·300	0·0	338,129	0	$x = 342,412$
	−2·191	1·239	0·650	329,537	64,900	$x = 344,111$
	−2·014	1·062	1·256	304,359	125,608	$x = 349,402$
6	−2·275	0·0871	−1·256	332,002	−121,900	$x = 184,319$
	−2·516	0·0224	−0·650	350,547	−62,297	$x = 162,973$
	−2·600	0·0	0·0	357,317	0	$x = 154,818$
	−2·516	0·0224	0·650	350,547	62,297	$x = 162,973$
	−2·275	0·0871	1·256	332,002	121,900	$x = 184,319$
7	−2·022	0·0163	−1·632	15,806	−257,553	$x = 80,859$
	−2·269	−0·4123	−1·202	12,242	−200,994	$x = 15,006$
	−2·388	−0·780	−0·672	−6000	−132,000	
	−2·372	−1·062	−0·0807	−29,000	−72,000	
	−2·222	−1·239	0·5326	−31,524	−18,914	$x = 4834$
8	−1·921	−0·4536	−1·691	16,094	−270,718	$x = 142,644$
	−1·932	−1·039	−1·396	−27,393	−235,611	$x = 97,522$
	−1·849	−1·541	−0·983	−69,432	−189,596	$x = 68,681$
	−1·678	−1·927	−0·4812	−108,993	−137,936	$x = 58,358$
	−1·432	−2·168	0·0761	−145,467	−83,300	$x = 67,869$

Table 2 (*continued*)

Point of ejection	Initial velocity (km/sec)			Value of y when $x = 0$ (km)	Value of z when $x = 0$ (km)	Remarks
	V_x	V_y	V_z			
9	−1·818	−0·1742	−1·85	99,525	−293,677	$x = 233,782$
	−1·921	0·4536	−1·691	159,883	−277,863	$x = 264,943$
	−1·932	1·039	−1·396	211,358	−247,834	$x = 302,777$
	−1·849	1·541	−0·983	249,351	−204,314	$x = 344,826$
	−1·678	1·927	−0·4812	271,479	−150,741	$x = 387,843$
11	−2·412	0·4758	−0·8435	137,478	−258,817	$x = 58,535$
	−2·568	0·0163	−0·4025	119,000	−186,500	
	−2·566	−0·4123	0·0938	64,500	−113,300	
	−2·404	−0·780	0·4124	38,000	−78,000	
	−2·095	−1·062	1·116	6400	−10,440	
12	−2·412	−0·4758	−0·8435	190,378	−265,545	$x = 106,669$
	−2·568	−0·0163	−0·4025	237,916	−211,010	$x = 95,886$
	−2·566	0·4123	0·0938	274,109	−153,082	$x = 106,398$
	−2·404	0·780	0·4124	281,519	−120,678	$x = 156,309$
	−2·095	1·062	1·116	297,930	−38,375	$x = 188,092$
13	−2·275	−1·256	0·0871	59,000	−143,800	
	−2·516	−0·65	0·0224	117,000	−154,600	
	−2·600	0·0	0·0	197,000	−172,000	
	−2·516	−0·65	0·0224	245,473	−183,285	$x = 60,272$
	−2·275	1·256	0·0871	281,605	−185,703	$x = 132,516$

* When a value of x is given in the column of Remarks, the y and z values correspond to this value of x, which is the last one computed for that orbit.

that the particles that hit the earth leave the moon within a very small solid angle (around 20 square degrees), whether they fall in a localized region or cover a whole hemisphere. This clearly indicates that although in some cases ejection of material from the moon could spread tektites over half the earth, they would be so thinly distributed that we would not expect to find them; in concentrated falls, however, the probability of finding the tektites would be increased by a factor equal to the ratio of the areas covered, since the total number of tektites falling is the same in both cases. Some tektites that miss the earth on their first passage near it may eventually fall on it. Their number, however, would be very small since the velocity of escape from the earth–moon system is only slightly higher than that from the moon itself, and therefore these tektites would be lost by the system. Furthermore, a fraction of those tektites which remain in the system would fall back on the moon, thus reducing further the number of isolated falls on the earth.

As a good example of a concentrated fall I analysed in detail the orbits of particles leaving from point 1. Table 3 lists the initial velocities and points of arrival, which are plotted on Fig. 4 and again on Fig. 5, where Australia is shown for comparison.

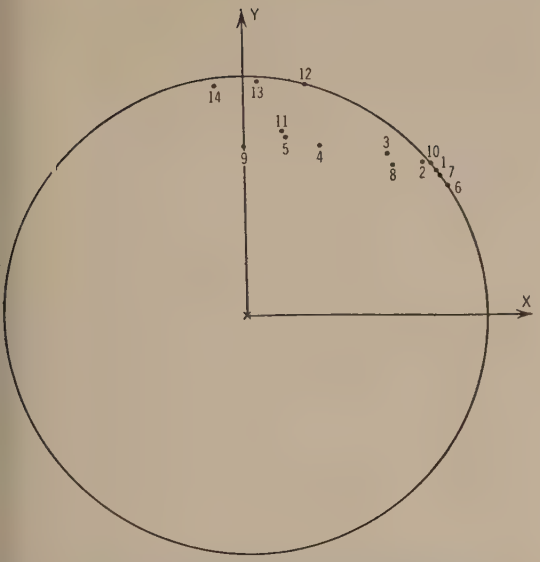

Fig. 4(a). An x–y projection of the points of arrival of tektites ejected from point 1 on the moon.

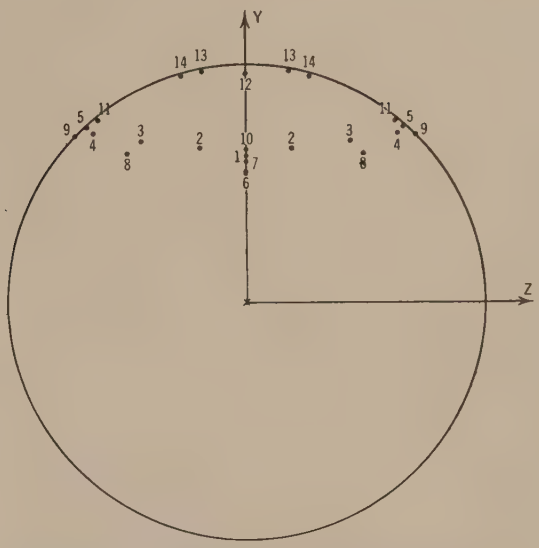

Fig. 4(b). A y–z projection of the points of arrival of tektites ejected from point 1 on the moon.

Fig. 5. The computed distribution of tektites ejected from point 1.

Table 3. A detailed survey of point of ejection 1

V (km/sec)	i	V_x (km/sec)	V_y (km/sec)	V_z (km/sec)	x (km)	y (km)	z (km)
2·400	15	−2·400	0·000	0·000			
2·450	15	−2·450	0·000	0·000			
	18	−2·447	0·128	0·000			
2·500	15	−2·500	0·000	0·000			
		−2·497	0·001	0·034			
		−2·491	0·002	0·059			
		−2·483	0·005	0·079			
		−2·471	0·008	0·101			
	17	−2·498	0·087	0·000			
		−2·495	0·088	0·038			
		−2·490	0·090	0·064			
2·510	15	−2·510	0·000	0·000			
		−2·474	0·010	0·421			
2·600	15	−2·600	0·000	0·000	5000	3950	0
		−2·599	0·000	0·047	4680	4150	1200
		−2·599	0·000	−0·047	4680	4150	−1200
		−2·596	0·001	0·131	3750	4350	2750
		−2·596	0·001	−0·131	3750	4350	−2750
		−2·594	0·002	0·175	1970	4550	4000
		−2·594	0·002	−0·175	1970	4550	−4000
		−2·586	0·004	0·262	1100	4750	4140
		−2·586	0·004	−0·262	1100	4750	−4140
		−2·582	0·005	0·306			
		−2·582	0·005	−0·306			
		−2·576	0·006	0·350			
		−2·576	0·006	−0·350			
		−2·570	0·008	0·393			
		−2·570	0·008	−0·393			
		−2·546	0·014	0·522			
		−2·546	0·014	−0·522			
		−2·563	0·010	0·436			
		−2·555	0·012	0·479			
2·600	16	−2·599	0·045	0·000			
		−2·596	0·046	0·131			
		−2·590	0·048	0·218			
		−2·582	0·050	0·305			
		−2·570	0·053	0·391			
	17	−2·599	0·091	0·0			
		−2·598	0·091	0·004			
		−2·595	0·092	0·132			
		−2·589	0·093	0·221			
		−2·581	0·096	0·299			
	18	−2·597	0·136	0·000			
		−2·595	0·136	0·086			

Table 3 (*continued*)

V (km/sec)	i	V_x (km/sec)	V_y (km/sec)	V_z (km/sec)	x (km)	y (km)	z (km)
		−2·591	0·138	0·166			
		−2·584	0·140	0·252			
2·7	15	−2·700	0·000	0·000	5280	3560	0
		−2·676	0·007	0·363			
		−2·676	0·007	−0·363			
2·8	15	−2·800	0·000	0·000	5100	3815	0
		−2·798	0·000	0·094	3865	4020	3080
		−2·798	0·000	−0·094	3865	4020	−3080
		−2·794	0·002	0·189	0	4500	4510
		−2·794	0·002	−0·189	0	4500	−4510
		−2·786	0·004	0·283			
		−2·786	0·004	−0·283			
		−2·775	0·007	0·376			
		−2·768	0·009	0·423			
		−2·760	0·011	0·470			
	16	−2·799	0·049	0·000			
		−2·796	0·050	0·141			
		−2·790	0·052	0·235			
		−2·780	0·054	0·328			
	17	−2·798	0·098	0·040			
		−2·797	0·098	0·084			
		−2·792	0·099	0·187			
		−2·784	0·101	0·281			
3·0	15	3·000	0·000	0·000	4900	4070	0
		−2·996	0·001	0·152	1000	4900	3945
		−2·996	0·001	−0·152	1000	4900	−3945
		−2·993	0·002	0·189			
		−2·993	0·002	0·189			
3·000	15	−2·989	0·003	0·253			
		−2·989	0·003	−0·253			
		−2·985	0·004	0·303			
		−2·965	0·009	0·453			
	16	−2·999	0·052	0·000			
		−2·996	0·053	0·151			
		−2·989	0·055	0·251			
		−2·984	0·056	0·301			
	17	−2·998	0·105	0·000			
		−2·996	0·105	0·114			
		−2·991	0·107	0·206			
3·200	15	−3·200	0·000	0·000	1600	6150	0
		−3·198	0·000	0·029	350	6250	1180
		−3·198	0	−0·029	350	6250	−1180
		−3·193	0·002	0·058	−750	6100	1675
		−3·193	0·002	−0·058	−750	6100	−1675
	17	−3·198	0·112	0·016			
		−3·198	0·112	−0·016			
		−3·194	0·113	0·160			
3·400	15	−3·400	0·000	0·000			
		−3·399	0·000	0·015			
		−3·399	0·000	−0·015			

Table 3 (*continued*)

V (km/sec)	i	V_x (km/sec)	V_y (km/sec)	V_z (km/sec)	x (km)	y (km)	z (km)
		−3·398	0·001	0·031			
		−3·398	0·001	−0·031			
		−3·395	0·001	0·172			
		−3·382	0·005	0·343			
		−3·361	0·010	0·514			
	16	−3·399	0·059	0·058			
		−3·399	0·059	−0·058			
	17	−3·398	0·119	0·000			
		−3·397	0·119	0·000			
3·500	15	−3·500	0·000	0·000			
		−3·499	0·000	0·016			
		−3·498	0·001	0·032			
	16	−3·499	0·061	0·000			
		−3·499	0·061	0·017			
3·600	15	−3·600	0·000	0·000			
		−3·599	0·000	0·016			
	16	−3·599	0·063	0·000			
3·700	15	−3·700	0·000	0·000			

V = magnitude of the velocity of ejection.
i = angle between the direction of ejection and the horizontal.

5. Conclusions

A comparison of Figs. 1 and 5 shows a strong similarity between the observed distribution of australites and the computed distribution of lunar debris. Such remarkable correspondence between the two lends very strong support to our model.

One striking feature about the distribution of australites is the fact that none are found north of an almost straight line, south of which they occur in large numbers. This feature comes about in the following way. The slowest particles that reach the earth fall on a line about 150 km south of the edge of the region of fall; faster particles fall farther north until a velocity of about 2·7 km/sec is reached; still faster particles fall again south of this edge, so that particles ejected with velocities of 2·8 km/sec and 2·6 km/sec fall at the same place. This means that no tektites should be found north of a certain sharp line, south of which a maximum concentration is to be expected. The concentration remains fairly constant for about 200 km in the north–south direction, and decreases to practically zero about 800 km south of the edge. In longitude, the concentration is a maximum at the centre of the region and decreases slowly towards the wings. Widely scattered tektites can be expected to fall at great distances from the region of high concentration, as shown in Fig. 5. Similar situations arise from points 14 and 15, although not enough orbits have been calculated for these points to determine so accurately the size and shape of the region of fall.

We can summarize the problem of the origin of tektites in the following way. Chemical evidence suggests that tektites may come from an extra-terrestrial

source, among which the moon is a possible one. Astronomical considerations prove that the moon is a very likely source compatible with the observed distribution of tektites on the earth's surface. Therefore we believe that these peculiar objects may well be, indeed, fragments of lunar material that reached our planet in the manner suggested in this paper.

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Achondrite investigations and their bearing on the origin of tektites*

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Abstract—A common origin for chondrites achondrites, and tektites seems reasonable. A lunar impactite origin for tektites seems doubtful because their content of cosmic-ray-induced ^{26}Al and ^{10}Be indicates much longer times of travel as small bodies in space than is possible in passing from the moon to the earth. A planet with a chondritic centre, an achondritic subcrust, and a tektitic crust is suggested as a possible place of origin. At the time of its disruption, the outer portions of the planet must have been hot enough so that the tektitic crust was molten and the achondritic sublayer was semi-molten. Achondrite compositions are examined from this viewpoint. With increasing SiO_2 the composition of the achondrite series varies as follows: FeO, CaO and Al_2O_3 undergo fractionations of 20 : 1, 6 : 1 and 7 : 1, respectively, while MgO is enriched by a factor of 8. This variation could not have been produced by crystal fractionation in a basic magma, but might have been produced by partition of chemical components between the achondrite and tektite layers due to a condition of liquid immiscibility in such a basic magma. This variation could also have been produced by crystal fractionation in an extremely acidic magma but the acidic magma itself would have had to have been produced by a segregation due to liquid immiscibility. Both methods of accounting for the observed compositional variations among achondrites would result in the formation of a siliceous phase similar in composition to tektites.

Introduction

Elsewhere (Cassidy, 1956) I have given reasons for believing that tektites are meteorites and have suggested a hypothesis that could explain their peculiar features. Basic assumptions of the hypothesis for their origin are (1) that tektite material was originally associated with other meteorite material as components of a planet, and (2) that this planet was disrupted to produce meteorites.

Because tektites occur on the earth within definite, limited areas, they must have entered the earth's atmosphere as clouds of small bodies. Varsavsky (1957) has stated that the possibility is very small that a cloud of tektites could travel any great distance through space without becoming dispersed. If this is true, a cloud of tektites could not travel much farther than the distance from the moon to the earth without spreading widely. He feels, therefore, that tektites must have been produced as spashes of melted rock from the impact of meteorites on the moon. The amounts of cosmic-ray-induced Be^{10} and Al^{26} found in tektites by Ehmann (1957) seem to indicate much longer times of exposure to cosmic radiation as small bodies than Varsavsky thinks is possible, however. For this reason the idea of a lunar origin for tektites must remain doubtful. From a spectroscopic analysis of trace elements in tektites and impactites, Preuss (1934) concluded that tektites are probably not terrestrial impactites. There seems to be no reason why his arguments should not apply to the question of lunar impactites also.

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Outline of the Hypothesis

Australian tektites (australites) are believed to be meteorites because their areal distribution suggests a meteoritic strewnfield and their "secondary" characteristics such as flanges, flow-ridges, and piezoglyph-like indentations suggest swift passage through the air. The australites are considered to have arrived in the earth's atmosphere as glassy bodies that already possessed rotational shapes such as spheroids, ellipsoids, dumbbells, and teardrops. If australites are meteorites, certain other kinds of tektites also are meteorites.

Given the previously described conditions of origin, tektites would roughly correspond to granitic material, as would stony meteorites to the basic mantle of the earth, and siderites to its high density core. Furthermore, just as the earth's surface may have been molten due to radioactive heating early in its history, the surface or near-surface layers of the meteorite planet must have been molten at the time of its disruption, in order to produce the tektite glasses. The low content of radioactive materials in stony meteorites and tektites need not cast doubt on the idea that melting would occur; Urry (1951) has pointed out that given enough time for heat accumulation, the major factor that determines whether or not melting will occur is the thickness of the radioactive-bearing layer and not the relative concentration of radioactive materials.

LaPaz (1958) has very recently presented strong evidence that some types of meteorites have come from outside the solar system. These include all siderites, pallasites and certain of the chondrites. The remainder are still assumed by him to have had their origin within the solar system. It has been suggested before (LaPaz, 1952) that some extraterrestrial material may originate outside the solar system. This additional evidence for the extra-solar system origin of some meteorite types now allows consideration of a much more simply constructed planet. The planet can now be thought of as composed of only three zones: *a chondritic centre, an achondritic subcrust, and a tektitic crust.* In its deepest part, the chondritic central portion would contain much nickel–iron, approximating the proportion found in mesosiderites. Thus, there would have been no metallic core. This is more in line with what would be expected in small planets like the moon and Mars.

To summarize the hypothesis, at the time of disruption the following conditions obtained at various levels within the planet. (1) The planet's central portion of chondritic material was at a temperature enough above its solidus so that small accumulations of liquid were present. Upon disruption of the planet and consequent sudden cooling, the liquid accumulations solidified to the glass plus radiating pyroxene crystals so commonly seen as chondrules. (2) The temperature of the achondritic subcrust was above its solidus but below its liquidus, so that the subcrust consisted of liquid plus crystals. Upon disruption of the planet the partly crystalline achondritic material suddenly cooled to produce aggregations of phenocrysts in a matrix of quench crystals. (3) The tektitic outer layer was above its liquidus temperature and thus was completely liquid. Upon disruption of the planet this liquid layer separated into clouds of droplets that adopted rotational shapes before quenching to a glass.

The temperatures of the achondritic and tektitic layers could have been the same before disruption, since the liquidus temperature of an achondritic melt is

somewhat higher than that of a tektitic melt. Therefore within a certain range of temperatures achondritic material would be partly liquid and partly crystalline while tektitic material would be all liquid.

In order to account for compositional differences between tektites and achondrites within the framework of this hypothesis it seemed necessary to suggest immiscible relations at the interface between the tektitic and achondritic melts. The purpose of the present paper is to call attention to the peculiar compositional trends among the achondrites and to point out that such a rock series cannot be produced by such recognized processes as crystal fractionation and segregation by filter pressing in a basic magma. If these processes cannot be called upon, the supposition of some process of segregation due to liquid immiscibility may be able to explain the achondrite rock series.

New Material

The new material presented in this paper is in the form of variation diagrams of stony meteorites and tektites. Figs. 1–6 are variation diagrams showing weight percentages of Na_2O, K_2O, FeO, CaO, Al_2O_3 and MgO plotted against SiO_2 for tektites and for stony meteorites. Chondrites, achondrites and tektites are included on the same diagrams because a common origin is being considered here. In this connexion, however, further justification for the association of chondrites and achondrites can be cited in the occurrence of chondritic inclusions in the Cumberland Falls achondrite as described by Merrill (1921). Originally these inclusions may have been xenoliths from the underlying chondritic mantle. In the construction of the diagrams all readily-available analyses were used, and are listed below. No attempt was made to evaluate analyses critically for the purpose of weeding out questionable ones; all were included. Since the diagrams were made before LaPaz's paper (1958) appeared, the chondrites plotted include many that LaPaz feels are of extra-solar system origin.

The following discussion considers those achondrite analyses lying between 45 and 60 per cent SiO_2. The chemical components of the chondrites show no discernible trends with increasing SiO_2, but definite trends are present within the achondrite group. Certain trends among the oxides which occur as a result of crystal fractionation in molten magmas have been cited by Bowen (1928) and others for many terrestrial rock series. These trends will now be contrasted with those found in achondrites. In the following sections, the trend of each oxide will be described first for terrestrial rock series containing 50–60 per cent SiO_2, and then for achondrites containing 45–60 per cent SiO_2.

Na_2O. In terrestrial igneous rocks Na_2O is fairly constant around 3 per cent over the range 50–60 per cent SiO_2, while in achondrites Na_2O is fairly constant around 1 per cent.

K_2O. In terrestrial igneous rocks K_2O is fairly constant around 1–2 per cent over this range, while in achondrites K_2O is quite constant at 0·5 per cent.

FeO, Fe_2O_3. In terrestrial igneous rocks Fe_2O_3 decreases steadily from 12 per cent to 7 per cent. (These figures are given for the sake of completeness but too much reliance should not be placed on them. It should be pointed out that Fe_2O_3 has apparently been enriched by fractional crystallization in some rock series.)

In achondrites, on the other hand, FeO decreases very sharply from 21 per cent to 1 per cent. (Fe_2O_3 is negligible because of the presence of free Fe).

CaO. In terrestrial igneous rocks CaO decreases regularly from 9 per cent to 7 per cent, while in achondrites CaO decreases very sharply from 12 per cent to 2 per cent.

Al_2O_3. In terrestrial igneous rocks Al_2O_3 can be considered constant between 16 and 17 per cent from 50 to 60 per cent SiO_2, but in achondrites Al_2O_3 decreases very sharply from 15 per cent to 2 per cent from 45 to 60 per cent SiO_2.

MgO. In terrestrial igneous rocks MgO decreases steadily from 6 per cent to 3 per cent, while in achondrites MgO increases sharply from 5 per cent to 40 per cent.

Discussion

Compositional variations among the achondrites will be discussed first from the premise that the parent magma was basic in composition, i.e. below about 60 per cent SiO_2. The variations will then be discussed from the premise that the parent magma contained enough SiO_2 so that it was in the primary phase field of SiO_2.

From the first premise, the grouping of the achondrite compositions along trends without extreme scattering is considered good enough to suggest an interrelated rock series that has not been affected too much by crystal settling. This is probably unfounded for the more basic achondrites, but it can be shown to be a valid assumption for the achondrites high in SiO_2, which are essentially monomineralic. Both phenocrysts and groundmass in these types consist of highly magnesian enstatite and the bulk composition is also that of highly magnesian enstatite. Thus, no matter whether the bulk composition or the composition of the interstitial material is considered, the result is a magma higher in both SiO_2 and MgO, and lower in CaO, FeO and Al_2O_3 than the more basic of the achondrites. Bowen (1928) has shown clearly that MgO is the first major oxide to disappear substantially from the residual liquid during a process of crystal fractionation in a magma containing less than about 60 per cent SiO_2, but in the achondrite series it is seen to increase steadily until the "residual liquid" is almost pure magnesian enstatite. Concurrently, CaO undergoes a fractionation of 6 : 1, FeO a fractionation of 20 : 1, and Al_2O_3 a fractionation of 7 : 1. This reduction in Al_2O_3 is as inexplicible by fractional crystallization processes as the remarkable increase in MgO with which it is associated. The sharp reduction in FeO and CaO with increasing SiO_2 is also quite unexpected. It seems logical therefore to suspect that a type of segregation completely different from crystal fractionation must be called upon to explain the trends in the achondrite series if the parent magma contained less than about 60 per cent SiO_2. Differentiation by segregation of a molten magma into two immiscible liquid phases is suggested, where the achondritic sublayer, at least near its top, was one phase, and the outer shell of tektitic material was the other. Due to immiscibility partitioning the composition of each phase would undergo marked changes near the interface, and these changes would not be the same as those produced in magmas by fractional crystallization.

Starting from the alternate premise that the parent magma contained enough SiO_2 so that it was in the primary phase field of SiO_2, the compositional trends among achondrites actually can be explained by a process of crystal settling. The

method of explanation is the same as that used by Bowen (1928), with reference to simple ternary phase equilibrium diagrams. Considering the MgO–FeO–SiO_2 and MgO–CaO–SiO_2 diagrams, and assuming the composition of the parent magma to lie within the primary phase field of SiO_2, so that cristobalite will be the first crystalline phase to appear in the melt, it can be shown that as more and more cristobalite crystallizes out of the melt, the composition of the residual liquid must move away from the SiO_2 apex and toward the pyroxene–cristobalite boundary. When this boundary is reached, pyroxene starts to crystallize from the melt and cristobalite continues to do so, so that the composition of the residual liquid moves along the pyroxene–cristobalite boundary, while the crystallizing pyroxene becomes richer in FeO and CaO and poorer in MgO. Cristobalite in the magma would not sink, and might rise, but the pyroxene crystals would probably sink and accumulate at the bottom of the molten layer. The first pyroxenes to accumulate could be highly magnesian if the parent magma were of the right composition, and successive increments of pyroxene crystals would be less and less magnesian and more and more calcic and ferrous. The result would be that samples taken from different levels within this zone of crystal accumulation would show highly magnesian enstatite near the bottom, grading toward ferrosilite and hedenbergite near the top, as more and more CaO and FeO appeared in the pyroxene. Toward the end of the crystallization process the residual liquid would largely be depleted in MgO and lower in SiO_2; olivine and calcic plagioclase would make their first appearances as crystalline phases. If the magma were allowed to congeal, the final result would be a surface layer of cristobalite, tridymite or quartz overlying a sublayer of settled crystals that graded downward from rock containing anorthite, olivine, and ferrous and calcic pyroxene near the top to magnesian enstatite near the bottom. If the compositional variations in the sublayer were then plotted on weight per cent diagrams such as those of Figs. 1–6, the weight per cent of SiO_2 would increase toward the base of the sublayer, since enstatite has a higher weight per cent SiO_2 than any other pyroxene. With increasing SiO_2 and increasing depth within the sublayer, the weight per cent MgO would increase and CaO, FeO and Al_2O_3 would drop steadily. These trends are the same as those observed among the achondrites. It should be noted in passing that according to this explanation, involving a very siliceous parent magma, a rock series can be produced with a crystallization sequence almost the reverse of that described by Bowen's reaction principle for basic magmas.

Reverting now to the basic assumption of the hypothesis, that material of chondritic, achondritic, and tektitic compositions formed the meteorite planet, it can be seen that any reasonable combination of each would produce a bulk composition that would not lie in the primary phase field of SiO_2. In order to produce segregation within the planet into chondrite, achondrite, and tektite zones, some sort of partition process must have operated. It has been shown that a process of crystal fractionation could have produced the compositional trends observed in the achondrites, leaving a residual liquid rich in SiO_2 corresponding to the tektites, only if the parent magma for the achondrites and tektites were within the primary phase field of SiO_2. Bowen (1928), however, has emphasized that crystal fractionation in a magma can never cause a residual liquid to cross the

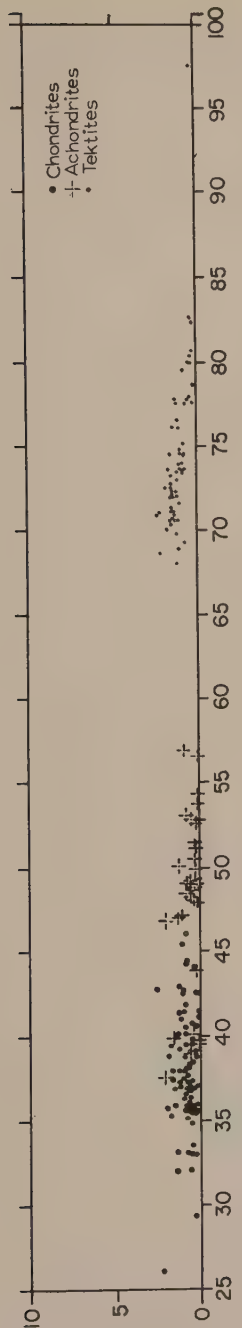

Fig. 1. Diagram showing the variation of Na_2O with increasing SiO_2 for chondrites (●), achondrites (+) and tektites (·).

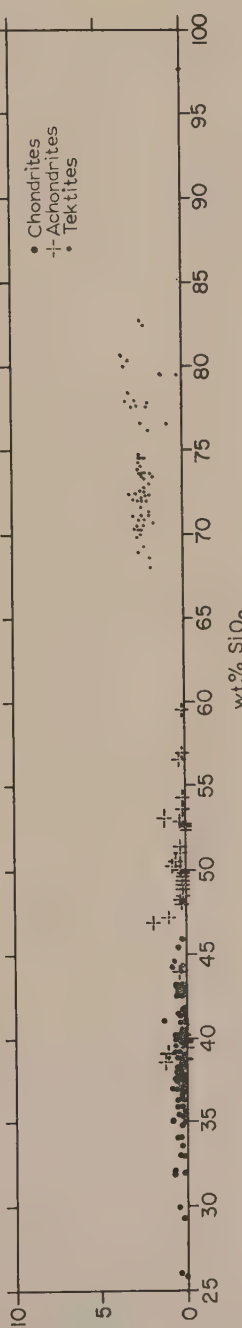

Fig. 2. Diagram showing the variation of K_2O with increasing SiO_2 for chondrites, achondrites and tektites.

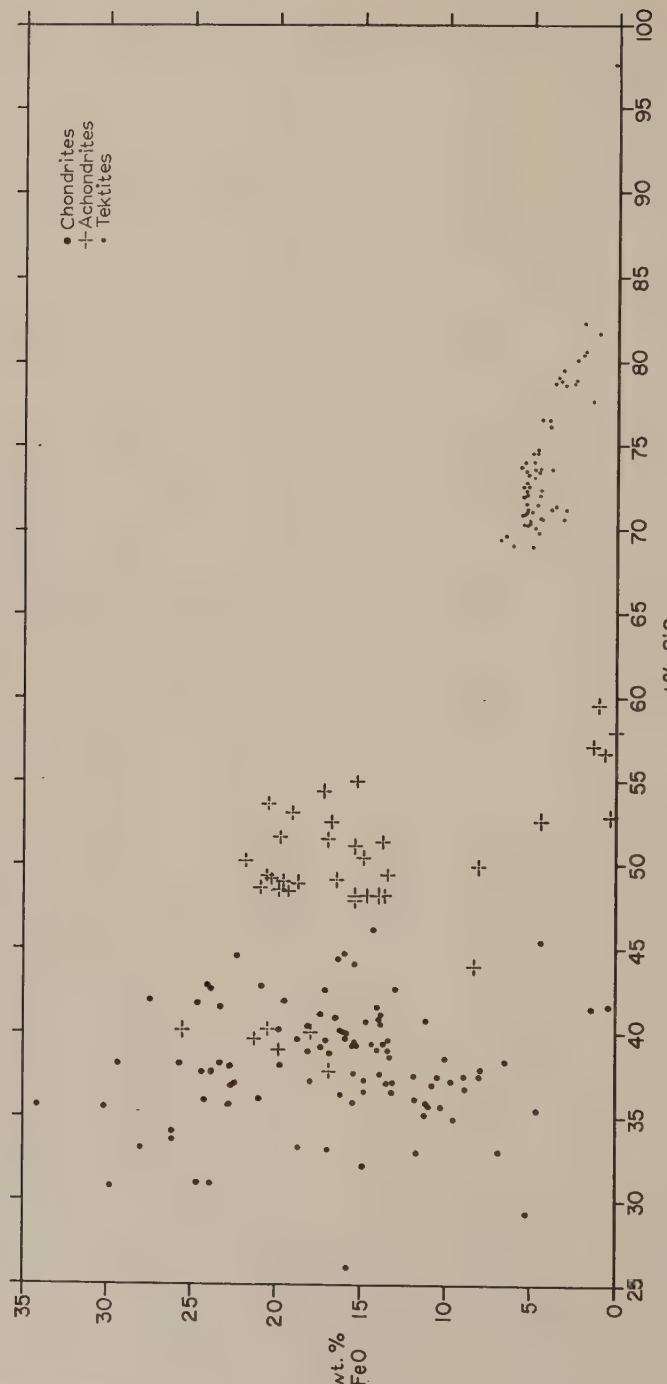

Fig. 3. Diagram showing the variation of FeO with increasing SiO_2 for chondrites, achondrites and tektites.

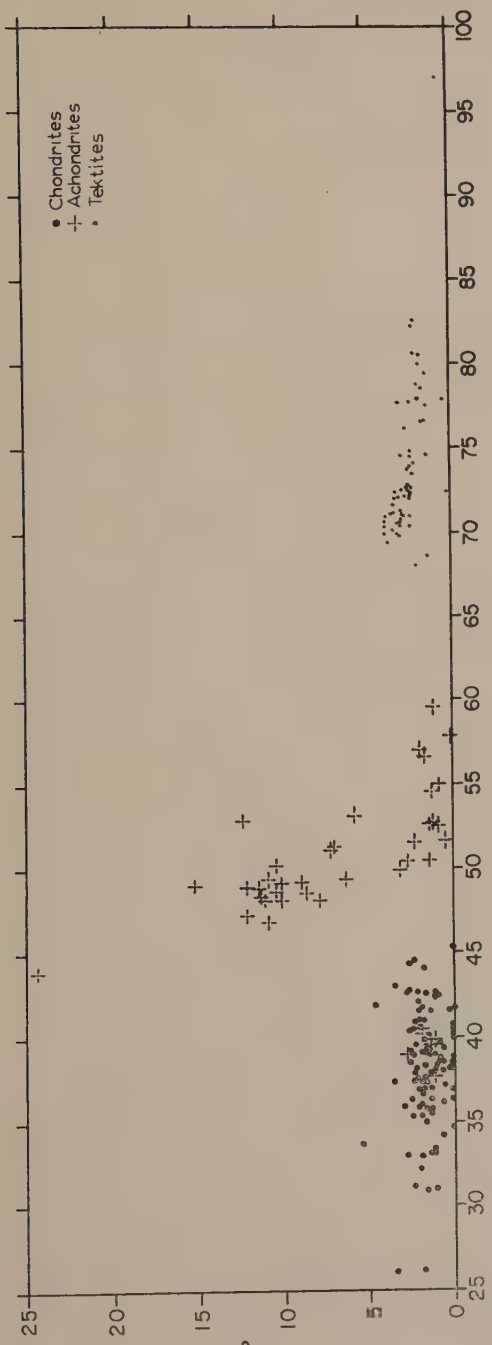

Fig. 4. Diagram showing the variation of CaO with increasing SiO_2 for chondrites, achondrites, and tektites.

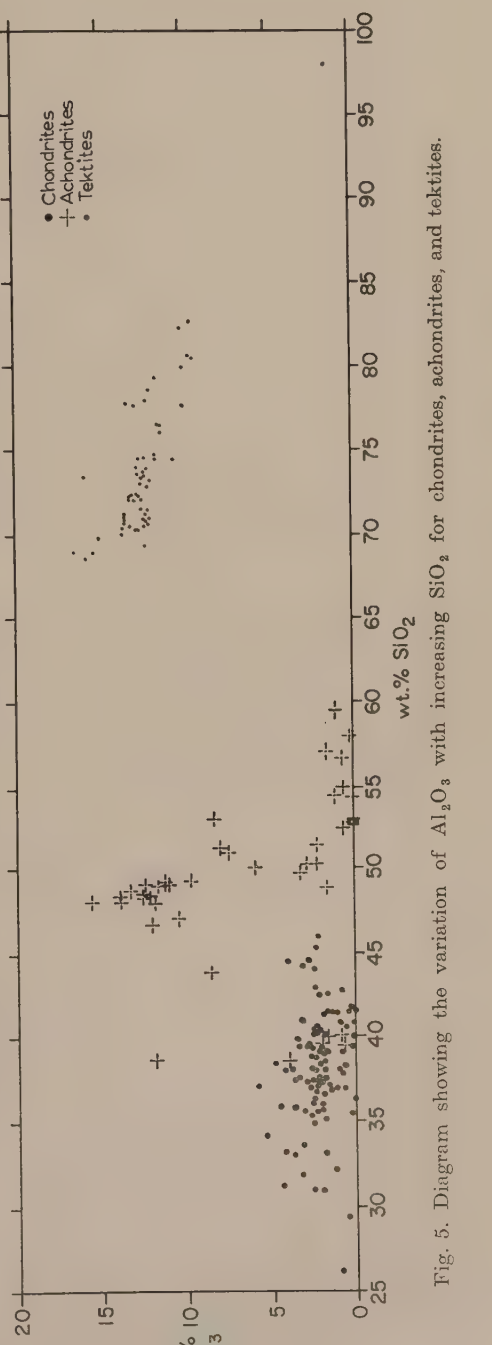

Fig. 5. Diagram showing the variation of Al_2O_3 with increasing SiO_2 for chondrites, achondrites, and tektites.

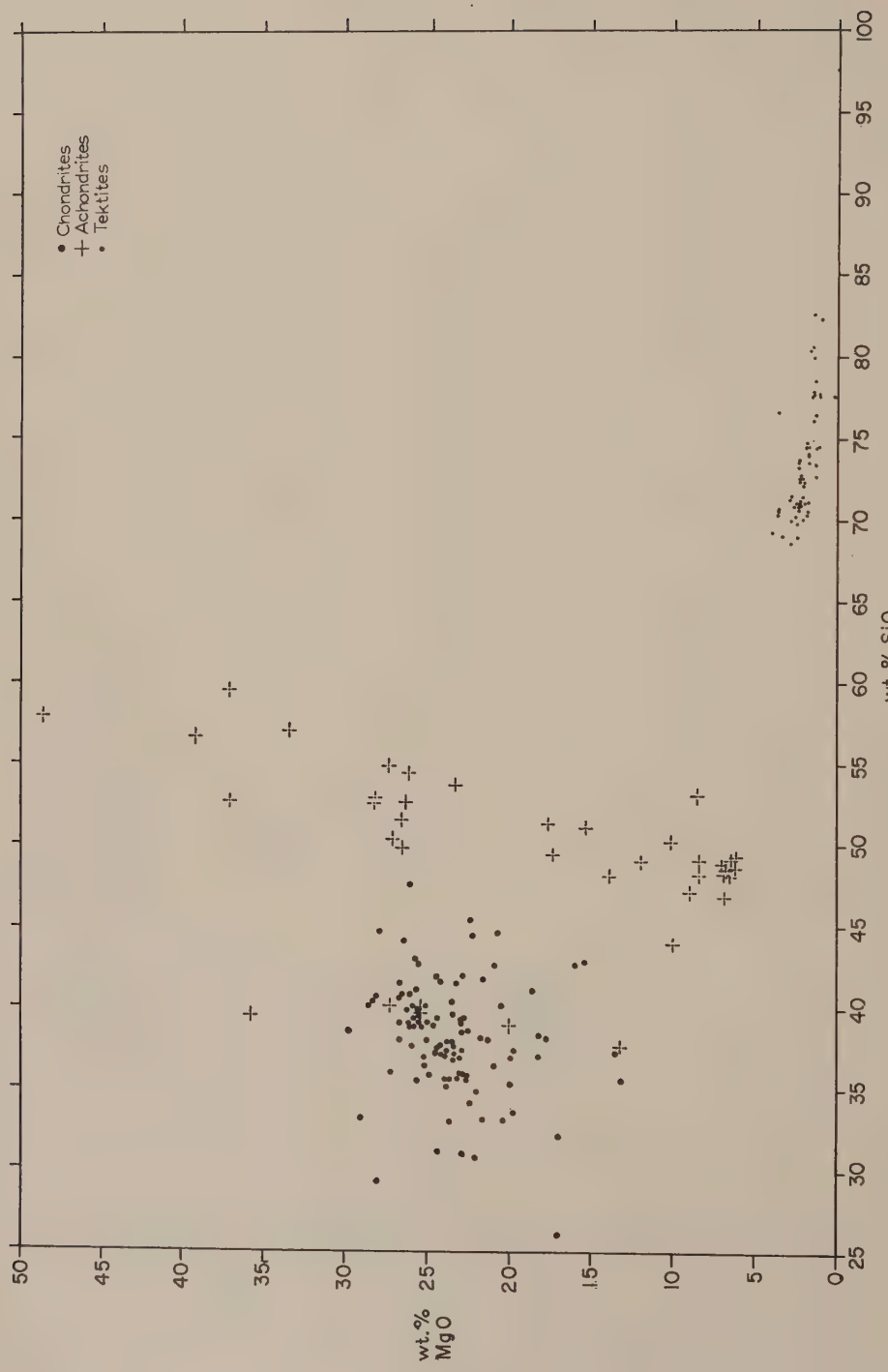

Fig. 6. Diagram showing the variation of MgO with increasing SiO_2 for chondrites, achondrites and tektites.

phase boundary into the primary phase field of SiO_2, so crystal fractionation could not produce, from the bulk composition of the meteorite planet, a residual liquid that could serve as a parent magma for achondrites. There can be no such limitation on processes of segregation by liquid immiscibility, however. A good example of this is the immiscibility region in the system K_2O–FeO–Al_2O_3–SiO_2 (Roedder, 1951), where the region of immiscibility extends into the primary phase fields of both SiO_2 and olivine. Therefore, under this alternate premise, in order to produce a highly siliceous parent magma for achondrites from a bulk composition consisting of chondrites, achondrites and tektites, segregation by liquid immiscibility is necessary.

In summary, the basic assumption has been made that chondrites, achondrites and tektites were originally associated as components of a planet in which partition had occurred to form a chondritic core, a tektitic crust, and an achondritic subcrust. The compositional variations within the achondrite rock series indicate that, had the parent magma of achondrites contained less than about 60 per cent SiO_2, the series could not have been derived by crystal fractionation. It is suggested that if the parent magma of achondrites did contain less than about 60 per cent SiO_2, the series might have been derived by some process of liquid immiscibility which would require the presence of a liquid of tektitic composition in equilibrium with a liquid of achondritic composition.

On the other hand, if the parental magma of the achondrites contained enough SiO_2 so that its composition lay in the primary phase field of SiO_2, crystal fractionation in the parent magma could produce the observed achondritic composition series if the achondrites are composed of crystals that settled to the bottom of the molten layer. It is pointed out that this would produce a rock series within the layer of settled crystals that would be higher in weight per cent SiO_2 at the bottom than at the top. An achondrite such as the Norton (Beck and LaPaz, 1951) would be found at the base of the layer and one such as the Moore County (Hess and Henderson, 1949) would be found near the top.

Since a parent magma lying in the primary phase field of SiO_2 could produce an achondritic layer and a tektitic layer by crystal fractionation, it seems logical to suspect that this is what happened. In order for this to have happened, however, this highly siliceous parent magma must have been derived from the bulk composition of the planet during partition of the layers within the planet. Since the bulk composition of the planet was undoubtedly too basic to lie within the primary phase field of SiO_2, the desired parent magma for achondrites could not have been produced by crystal fractionation but may have been produced by the formation of two immiscible liquids. If this were true, one liquid would be the parent magma of the achondrites and tektites while the other would be of chondritic composition.

It should be emphasized that all conclusions reached here have been based on the observed variations in composition among the achondrites, which are known to be meteorites. Both processes suggested here as possible methods for the production of the achondrite rock series require the operation of segregation due to liquid immiscibility and both processes would, by their operation, produce material of tektite composition in addition to the achondrite series. While the basic assumption called for an original association of chondrites, achondrites and

tektites, it was not necessary to assume the presence of tektite material since its production is automatic in both processes.

Experimental confirmation of the existence of a region of liquid immiscibility relating to meteorite compositions would provide strong support for the speculations made here. There are certain obvious difficulties inherent in such experimental work, not the least of which is the necessity of dealing with seven or eight components, but investigations in this line are being attempted.

Acknowledgement—Data for this paper were gathered and the paper was written under the sponsorship of a predoctoral fellowship from the National Science Foundation, for which I am duly grateful.

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Chondrites

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12. Padvarninkai: *M. A.* **4,** 419–20.
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The water, deuterium, gas and uranium content of tektites*

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Abstract—The water content, deuterium concentration of the water, total gas and uranium contents were determined on tektite samples and other glass samples from Texas, Australia, Philippine Islands, Java, French Indo-China, Czechoslovakia, Libyan Desert, Billiton Island, Thailand, French West Africa, Peru, and New Mexico. The water content ranges from 0·24 per cent for the Peru tektite, to 0·0002 per cent for a moldavite. The majority of the tektites have less than 0·05 per cent water, and average 0·005 per cent H_2O by weight. No other gases were detected, the lower detection limit being about 1 p.p.m. by weight. The deuterium content of the water in tektites is in the same range as that in terrestrial waters, and varies from 0·010 mole per cent to 0·0166 mole per cent deuterium. The uranium content is about from 1 to 3 p.p.m.

The possible origin of tektites is discussed. The experimental data presented favour their being originally terrestrial, but produced by some catastrophic event. An extra-terrestrial source is not ruled out.

Introduction

If the literature (Barnes, 1940) on the analysis of tektites is examined, one is immediately struck by an obvious inconsistency. The water content (H_2O—) of these objects as determined by the usual methods of rock analysis has been reported as between several hundredths per cent by weight, up to about one-tenth per cent. The analysis of gases extracted by heating tektites shows an absence of water and a predominance of carbon monoxide, carbon dioxide, and hydrogen.

The author's experience (Friedman and Smith, 1958) in the extraction of water from obsidians and other natural glasses suggested that the water content of tektites, as reported by chemical analysis, is high, owing mainly to contamination occasioned by grinding of the sample before analysis. A consideration of the methods used in the previous extraction of gases from tektites indicated that most of the H_2, CO_2 and CO was present in the tektite originally as water, and that these gases resulted from the reaction of water with carbon present in the sample container.

It was felt that an analysis of the water and its deuterium concentration, and of the gas and uranium content of these peculiar natural glasses would prove interesting.

Experimental Procedure

The surfaces of the tektites were first cleaned by sandblasting with coarse carborundum powder. The samples were then cut into suitable sized pieces with a water-cooled diamond saw. The surface was then removed by dry-grinding with a diamond loaded steel disk. Organic matter was dissolved by washing with CCl_4.

An 18 ml platinum J. Laurence Smith crucible was heated with a strongly oxidizing oxy–natural gas flame to 1000-1200°C for several minutes. The crucible and cover were then placed in a vacuum and heated by means of an induction heater to 1450°C for several minutes. After allowing the crucible to cool, the

* Publication authorized by the Director, U.S. Geological Survey.

sample (usually consisting of one or two pieces totalling approximately 5–10 g) was placed in the degassed crucible, the platinum cover placed over the crucible, and the whole resealed in the vacuum system (Fig. 1). The sample was evacuated to less than 10^{-4} mm of Hg, and heated to approximately 100–200°C for 15–30 min. During this period the crucible was "flashed", quickly heated to 800–900°C, for several seconds to degas. After checking for removal of adsorbed gas, the sample was isolated from the vacuum system, the trap (*C*) cooled with liquid nitrogen, and the crucible and sample quickly heated to approximately 600°C. The heat

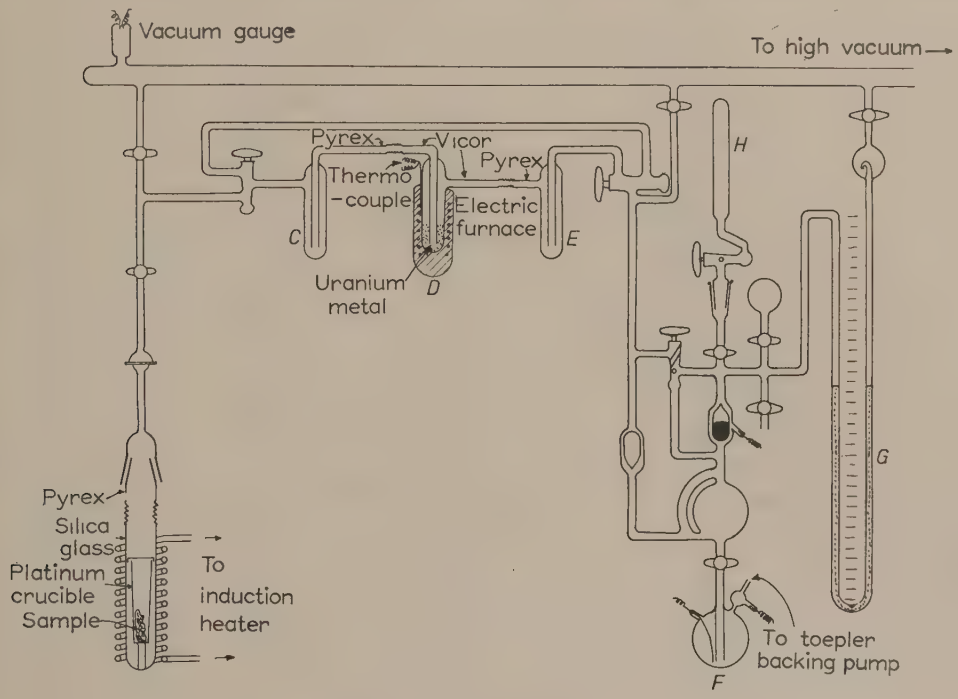

Fig. 1. Sample preparation line.

was slowly increased to reach 1450°C in about 30 min. It was kept at this temperature for 10 min.

To check for the presence of hydrogen, the Toepler pump was operated for a few minutes. In no case was any gas detected at this point, proving that less than 0·01 cm^3 S.T.P. of free hydrogen or other noncondensible gas was present in the samples. The trap (*C*) was then isolated from the sample, the liquid N_2 removed from around it, and trap (*E*) cooled with liquid N_2. The water that was collected in trap (*C*) then reacted with the hot uranium metal in furnace (*D*) to produce uranium oxide (solid) and hydrogen. The hydrogen was pumped into a calibrated manometer, and its pressure measured. It was then transferred to a mass spectrometer, and its D/H ratio determined. The precision of the water content measurement varies with the size and water content of the sample, but in general the results are reproducible to within 20 per cent of the amount present. Contamination will tend to make the results high, but loss of water by adsorption in the

equipment will probably compensate for this error. Occasional blanks yielded barely detectable amounts of water, and errors from this source are insignificant.

The D/H determinations were fairly inaccurate owing to the small amount of sample, and the possibility of exchange of the sample with hydrogen adsorbed in the mass spectrometer inlet system and source. A precision of $\pm 0{\cdot}0015$ mole per cent D is probably valid.

Table 1. Water content, relative deuterium concentration, and uranium content of tektites

I.F. sample no.	Locality	H_2O (%)	Mole % D	U (p.p.m.)
43–5	Libyan Desert	0·064	0·0143	1·1*
43–6	Philippines, Rizal	0·002	0·0144	2·7*
43–3	Philippines, Rizal	0·005	0·0143	2·7*
42–1	Philippines, Busuanga	0·004	0·0132	2·6*
96–6	Philippines, Balacan	0·002	0·0156	1·7†
96–2	Philippines, Luzon	0·014	0·0157	1·8†
43–4	Perth, Australia	0·007	0·0143	2·4*
97–2	Australia	0·005	0·0148	1·6†
42–2	French Indo-China	0·007	0·0140	2·4*
40–15	French Indo-China	0·005	0·0152	2·5*
96–10	French Indo-China	0·005	0·0146	1·1*
43–7	Texas	0·005	0·0148	2·4*
27–17	Texas	0·002	0·0146	1·1†
97–3	Bohemia	0·0005	—	1·6†
97–9	Bohemia	0·0003	—	1·5†
96–11	Java	0·002	0·0148	1·5†
96–7	Billiton Island	0·004	0·0151	1·4†
41–15	Peru	0·23	0·0127	18·0*
96–9	Thailand	0·01	0·0166	3·0†
93–4	Atom bomb, New Mexico	0·05	—	—
97–1	French West Africa	0·02	0·0140	0·91†
98–5	Beer bottle	0·02	—	—

Uranium determinations made two years apart:
* First batch, Roberta S. Dymond, Analyst, U.S. Geological Survey.
† Second batch, Roosevelt Moore, Analyst, U.S. Geological Survey.

The fluorometric uranium analyses (Grimaldi *et al.*, 1952) were done in two batches, two years apart. The results within each batch are probably precise to ± 10 per cent of the amount present, but changes in standards between the time the first and the second batch were analysed are such that the variation between batches may be as large as ± 50 per cent of the amount present.

Results

The results of this research are given in Table 1. Fig. 2 shows the deuterium concentration of ocean water, rain, snow, water in glassy rhyolitic rocks, and water in the tektites reported in this paper. The deuterium concentration of water in the tektites falls within the range of terrestrial materials.

This, then, does not preclude a terrestrial origin for these objects. Although

the cosmic abundance of deuterium is low, it is not impossible that other cosmic bodies could concentrate deuterium as has the earth. If tektites are formed from such bodies, the high abundance of deuterium in the tektites relative to the "cosmic abundance" can be explained. However, the similarity of deuterium values

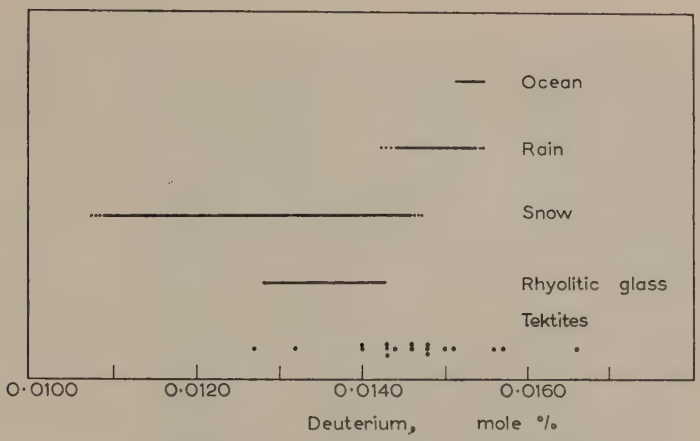

Fig. 2. Relative deuterium content of natural waters.

to normal terrestrial waters can be construed as evidence for their terrestrial origin.

The low water content of the tektites, however, is impressive (Fig. 3). They are obviously more anhydrous than any glass that we know was fused by natural processes on the earth. The glass formed by fusion below the fireball of an atom bomb approach the tektites in anhydrousness, as does the Aouelloul impactite glass from Adrar, French West Africa. As has been suspected before, the americanites are probably not true tektites, but are probably allied to volcanic glasses. The numerous large euhedral crystals present in the americanites also differentiate them from the rest of the tektites.

The Libyan Desert glass also seems to be different from the tektites. The water extracted from these samples represents a lower limit of the amount actually present, as the high silica content (~98 per cent SiO_2) of these samples results in the material having a melting point above the 1450°C reached in these experiments.

The Thailand glass (no. 96–9) is high in both water and uranium content. It is possible that this is also a terrestrially formed volcanic glass.

The remaining tektite samples contain less than 0·014 per cent H_2O, the majority less than 0·007 per cent. Their uranium content seems to be fairly constant, approximately 1–3 p.p.m.

The low water content of these glasses makes it highly probable that they were either melted in an environment whose water vapour pressure was several orders of magnitude lower than that usually experienced in terrestrial environments, or that they were heated to a very high temperature, in the order of 2000°C or above.

An interesting corollary of these experiments is that tektites containing as little as 20 p.p.m. of water vesiculated when heated as previously described. Fig. 4 shows a sample that originally contained 20 p.p.m. of water. The sample expanded by a factor of 5–8. Samples containing as little as 2 p.p.m. of water

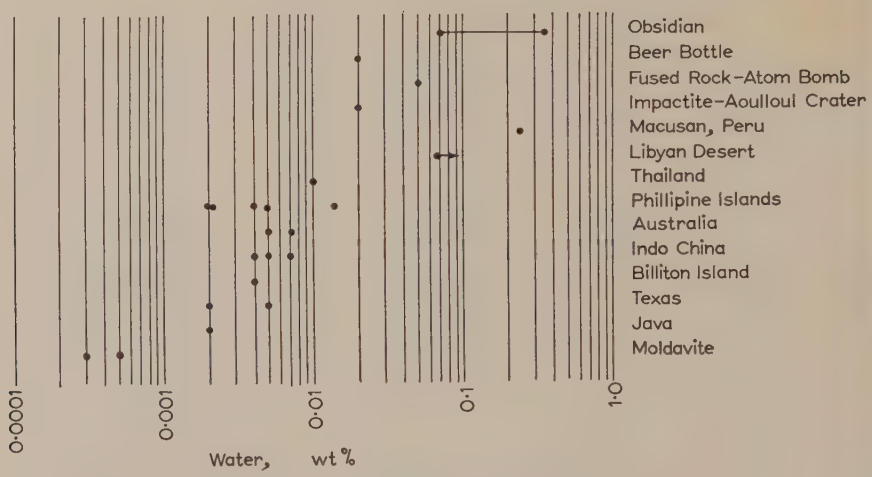

Fig. 3. Water content of tektites and other glasses.

showed a very slight frothing. The bubbles found in some tektites could have been formed by expansion of water during rapid heating in a dry environment. This is in accord with the results found by Suess (1951) who reported that the interior of the bubbles are at a pressure of less than 10^{-3} mm of mercury.

Unfortunately, the data presented in this paper do not in themselves allow the choice of a particular origin for tektites, but give additional information that has to be fitted in to any consistent theory of tektite origin. For example, both Urey's (1957) hypothesis of comet-explosion, and the lunar splash hypothesis will fit the experimental facts presented herein.

Urey postulates a comet, or comets, colliding with the earth's atmosphere. The resulting explosive shock-wave strikes the earth's surface, forming a large shallow crater, and melting large amounts of surface rock. These melted rocks may be thrown into the outer atmosphere, chilled, and partially remelted upon their fall back to the earth's surface.

This process could produce glasses that contained water whose deuterium concentration was similar to terrestrial waters, but whose water content was low owing to the high temperature of melting. Whether the high temperature could be maintained long enough to homogenize the glass and to allow most of the water to leave, and "fine" the glass (remove air and gas bubbles) is debatable. The difficulty of removing water may have restricted tektite formation by Urey's process to initially dry sediments found on a desert surface. If the sediments are heated rapidly to 2000–3000°C, the escaping steam will cause an explosion. If they are heated slowly, vesiculation will probably occur, and a long period of subsequent heating at high temperature would be required to remove the bubbles.

Fig. 4. Photograph of crucible containing tektite after fusion and vesiculation.

The lunar splash origin for tektites is not ruled out by the deuterium and water results. One would have to postulate that the moon was able to accumulate and retain a small amount of water of deuterium concentration similar to that collected by the earth. Lunar material would have had to have been chemically differentiated in order to produce materials of tektitic composition with a high uranium content.

All suggested hypotheses of the origin of tektites contain difficulties. The author is of the opinion that tektites were produced from earth materials in a catastrophic event. Perhaps some modification or extension of UREY's hypothesis will be the answer.

Although no known geologic process can explain the formation of tektites, the similarity of tektites to terrestrial materials in bulk chemical composition, uranium and deuterium content, and in the isotopic composition of their lead content is too striking to be coincidental. We must, therefore, look for an unusual event to produce them.

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DESCRIPTION OF SAMPLES

I.F. no.	*Locality*
43–5	Libyan Desert glass donated by CLIFFORD FRONDEL (Harvard Museum) samples cut from centre of 200 g piece.
43–6	Rizalite, approximately 25 g donated by CLIFFORD FRONDEL, sample contains many small bubbles.
43–3	Rizalite, Santa Mesa, Rizal Province, P. I. National Museum no. 1461.
42–1	Busuanga, P. I., H. Otley Beyer no. 6, Kalervo Rankama analysis no. 61.
96–6	Bulacan, P. I., American Museum of Natural History no. 3790 weight = 33·7 g, $d = 2{\cdot}45$.
96–2	Coco Grove, Paracale, Camarines Norte, Luzon, P. I. donated by J. W. PEOPLES 45·0 g, $d = 2{\cdot}43$.
43–4	Australite, 75 miles east of Perth, western Australia, contained 4 mm diameter bubble, National Museum no. 104913.
97–2	Australite, collected near R. R. line from Abminga to Finke, south Australia by W. A. CASSIDY, weight = 42·3 g, $d = 2{\cdot}46$.

42–2	Indochinite, Tan-Hai, French Indo-China, Orcel (Paris) no. 6 Kalervo Rankama analysis no. 31.
40–15	North Cambodia, French Indo-China, Harvard Museum no. 92701, weight = 33 g.
96–10	Lang Bian, Annam, French Indo-China, American Museum of Natural History no. T–5, weight = 17 g, d = 2·43.
43–7	Bediasite, University of Texas Museum no. 30773, weight = 44 g.
27–17	Bediasite, University of Texas Museum no. 30763, weight = 35·1 g.
97–3	Moldavite, Habri in S. Bohemia, donated by Natural History Museum of Vienna, weight = 12·6 g, d = 2·37.
97–9	Moldavite, Nestomice, near Budweis, Bohemia, American Museum of Natural History no. T–15, weight = 22·3 g, d = 2·37.
96–11	Trivil beds, Kalrosso, Central Java, American Museum of Natural History no. T–9, weight = 22·4 g, d = 2·46.
96–7	Billitonite, Billiton Island, Netherlands East Indies, Harvard Museum no. 91367, weight = 19·1 g, d = 2·45.
41–15	Americanite, Macusani Prov. Carabaya Dept., Puno, Peru, Kalervo Rankama analysis no. 160.
96–9	Fang-Dang, Thailand (indochinite) collected by W. A. Cassidy.
93–4	Glassy melted rock from atom bomb crater Alamogordo, New Mexico, collected by Clarence Ross.
97–1	Impactite, glass from Aouelloul crater, Adrar, French West Africa, collected by W. A. Cassidy, courtesy Institute Francais d'Afrique Noire and the Institute of Meteoritics, University of New Mexico, weight = 96·1 g.
98–5	Beer bottle, Valley Forge beer, U.S.A.

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Isotopic composition of lead from tektites

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Abstract—The isotopic composition of lead in three tektites and Libyan Desert glass is compared with that in known terrestrial and extra-terrestrial sources. The lead contained in the glasses is similar to modern terrestrial lead, particularly lead from modern oceanic sediments. The uranium, thorium and lead concentrations were determined for one of the glasses, an australite. Evidence is given which indicates that within the last tens of millions of years differentiation of uranium, thorium and lead occurred in the parent material of the australite. These results are difficult to explain in terms of any extra-terrestrial origin involving fusion of materials from the moon, meteorites or comets, but they are readily explained if tektites are of terrestrial origin involving fusion of argillaceous sediments in some unspecified way.

Introduction

The isotopic compositions of modern terrestrial lead isolated from various sources such as young basalt, oceanic clay, and galenas (excluding "anomalous" galenas) are quite similar. The narrow range of isotopic composition (see Table 3 below) indicates that the lead was produced in environments having nearly equal uranium–lead and thorium–lead ratios. Moreover, information obtained from studies of the isotopic composition of galena lead as a function of time indicates that there has been little, if any, chemical separation of uranium and thorium from lead in the parent material of galenas during the past two or three billion years. Since average terrestrial leads are to be associated with closely defined ratios of uranium and thorium to lead, extra-terrestrial bodies which have had different proportions of these elements over periods of time which are of the order of the age of the solar system will yield lead of unusual isotopic composition. An example of this is the lead found in the Nuevo Laredo stone meteorite by Patterson (1955).

Since tektites have been considered to have a possible extra-terrestrial origin, an examination of the isotopic composition of their lead appeared profitable. Accordingly, lead was isolated from three tektites and a sample of Libyan Desert glass, analysed isotopically, and found to be similar to modern terrestrial lead in composition. The concentrations of uranium, thorium and lead were then determined for one of the glasses in order to see whether the uranium–lead and thorium–lead ratios had been changed recently, as might be expected if the source material was a terrestrial sediment. These studies constitute a preliminary investigation into the natural glass problem by the lead method, but the results are reported here since they provide information in an area where data have been completely lacking.

Experimental

The glasses used were supplied to the author by I. Friedman of the U.S. Geological Survey. The following descriptions accompanied the samples:

Australite: Collected near railroad line from Abminga to Finke, South Australia, by W. A. Cassidy. $d = 2{\cdot}46$; no. 97-2.

Moldavite: Habri in S. Bohemia. Courtesy of the Natural History Museum of Vienna. $d = 2{\cdot}37$; no. 97-3.

Philippinite: Bulacan. American Museum of Natural History sample no. 3790. $d = 2{\cdot}45$.

Libyan Desert glass: Courtesy of L. J. Spencer. Described by Spencer (1939).

The surfaces of the glasses had been sandblasted by Friedman. The samples were washed for several minutes in hot 6 M hydrochloric acid, rinsed in lead-free distilled water, and dried. They were then crushed in a steel mortar which had been used only for grinding basalts and ultra-basic rocks. The powders were decomposed in platinum dishes with hydrofluoric and perchloric acids. Lead was recovered by dithizone extractions, first at a pH of 8·5 in the presence of citrate and then at a pH of 9 in the presence of potassium cyanide. The samples yielded from 15 to 20 μg of lead, which may be compared to blanks of from 0·2 to 0·3 μg as measured by isotope dilution. The lead, uranium and thorium concentrations in the australite were all determined by the method of isotope dilution and are thought to be accurate within ± 1 per cent. These procedures have been described in previous papers (Tilton, Aldrich and Inghram, 1954; Tilton *et al.*, 1955; Aldrich *et al.*, 1956). Mass spectrometer runs were made by surface ionization using an instrument equipped with an electron multiplier.

In general, the glasses were nearly ideal materials as far as the analytical methods were concerned. They dissolved easily and the major constituent, silica, was removed during the decomposition. This permitted the use of very dilute solutions for subsequent chemical operations, leaving little danger of interferences from other elements during the extractions. The analyses are by no means as difficult as those for basalts or meteorites.

Results

Tables 1 and 2 give the isotopic composition of lead from the four glasses and the uranium, thorium and lead concentrations for the australite. The ratios in Table 1 are believed to have absolute errors of $\pm 0{\cdot}5$ per cent with regard to instrumental uncertainties. This is based on replicate analyses of two standard leads made up from mixtures of gravimetrically standardized solutions of nearly pure Pb^{206} and Pb^{208}. The standards have Pb^{206}/Pb^{208} ratios of approximately 1 and 20 in order to determine possible non-linearities in the mass spectrometer. It has so far been unnecessary to apply either electron multiplier or non-linearity corrections to the lead ratios. A more complete report of this work is planned for future publication. The ratios in Table 1 are based on Pb^{206} rather than on Pb^{204} as has generally been done in the past. The new method is a more accurate way of presenting the data since it bases the results on a high intensity peak of intermediate mass rather than on a low intensity peak of lowest mass. A Pb^{206}/Pb^{207} ratio can nearly always be measured more accurately than a Pb^{207}/Pb^{204} ratio, for instance, yet the Pb^{206}/Pb^{204} and Pb^{206}/Pb^{207} ratios for a lead contain the same essential information as do the Pb^{206}/Pb^{204} and Pb^{207}/Pb^{204} ratios.

The concentration values in Table 2 are believed to have analytical errors of ± 2 per cent for lead and ± 1 per cent for uranium and thorium. The lead result

Table 1. Isotopic composition of lead from some natural glasses

Source	Pb^{206}/Pb^{204}	Pb^{206}/Pb^{207}	Pb^{206}/Pb^{208}
Australite	18·81	1·206	0·486
Moldavite	18·55	1·185	0·483
Philippinite	18·83	1·199	0·482
Libyan Desert glass	19·11	1·213	0·488

Table 2. Concentrations of uranium, thorium and lead in the australite

Element	Amount taken for analysis (g)	Concentration (p.p.m.)
Uranium	0·587	1·74
Thorium	0·587	9·19
Lead	2·346	2·90

is corrected for a blank of 0·25 μg, uranium for a blank of 0·007 μg, and thorium for 0·02 μg. It is possible to estimate the lead concentrations in the moldavite, philippinite, and Libyan Desert glass from the amounts of dithizone which were required for the final lead extractions. An estimate of this type made for the australite agreed with the final isotope dilution value within 10 per cent. The following estimates are probably accurate within $\pm$20 per cent: moldavite, 6 p.p.m.; philippinite, 5 p.p.m.; Libyan Desert glass, 5 p.p.m.

Discussion

The data obtained on the glasses are in complete harmony with a terrestrial origin. First, the isotopic composition of the lead is similar to that of modern terrestrial lead. The leads in Table 1 are quite similar to each other although small differences probably exist. In particular the Libyan desert glass appears to have more Pb^{206} with respect to Pb^{204} and Pb^{207} than do the other glasses. Replicate determinations would be necessary in order to establish these differences with certainty. With the exception of the australite the samples were completely consumed in the analytical work reported in Tables 1 and 2. For purposes of comparison the isotopic compositions of some modern terrestrial leads are given in Table 3. The similarity of the leads in Table 1 to those in Table 3 is obvious. The leads from the glasses are particularly similar to those found in the Pacific Ocean clay and the manganese nodule. The discussions of this paper will ignore "anomalous" leads, that is, leads which have been associated with an unusually large amount of uranium (and sometimes thorium as well) with respect to lead for periods of time which are short with respect to the age of the earth. Anomalous leads often occur in known uraniferous provinces, for example.

A second similarity to terrestrial materials concerns the uranium–thorium–lead balance observed for the australite. That is, the relative amounts of uranium and thorium with respect to lead found in the glass can be compared with the uranium–lead and thorium–lead ratios calculated for the environments which have produced modern terrestrial lead. This comparison will yield information

Table 3. Isotopic compositions of some modern terrestrial leads

Source of lead	Reference	Pb^{206}/Pb^{204}	Pb^{206}/Pb^{207}	Pb^{206}/Pb^{208}
Snake River basalt	Patterson *et al.*, 1955	18·12	1·173	0·476
Red Clay, Pacific Ocean	Patterson *et al.*, 1955	18·95	1·202	0·487
Manganese nodule, Pacific Ocean	Patterson *et al.*, 1955	18·91	1·205	0·489
Fumarolic magnetite, Valley of Ten Thousand Smokes, Alaska	This paper	18·99	1·221	0·493
Average modern galena	Bate, 1955	18·9	1·19	0·482

concerning possible recent changes in the ratios of thorium to lead and uranium to lead which might have occurred in the source material of the australite. There are two ways in which the uranium–lead and thorium–lead ratios which have governed the evolution of terrestrial lead may be calculated. The first is to plot the isotopic composition of lead from galena deposits as a function of their age. Studies of this type indicate that galena leads have been produced in environments which have a U^{238}/Pb^{204} atom ratio of 9·8 and a Th^{232}/U^{238} ratio of 3·9 at the present time (Collins, Russell and Farquhar, 1953). As mentioned previously the observed variation of the isotopic composition of the leads is very nearly that which would be calculated for a source in which no differentiation of uranium, thorium and lead by processes other than radioactive decay have taken place.

A second method of calculating the uranium–lead and thorium–lead ratios of the source material which has produced terrestrial leads is to assume that the earth originated 4·6 billion years ago with lead of the same isotopic composition as that found in Canyon Diablo and Henbury troilites by Patterson (1956). Patterson has discussed the plausibility of this model in some detail and no further treatment will be given here. Using this model together with the isotopic composition of lead for an average modern galena as given in Table 3, the following atom ratios are calculated for the present-day environment which has produced the lead: $U^{238}/Pb^{204} = 9{\cdot}3$ and $Th^{232}/U^{238} = 4{\cdot}1$. These values are in quite good agreement with those calculated from the observed variation of the isotopic composition of lead in galenas with time.

Table 2 gives the concentrations of uranium, thorium and lead observed in the australite. From this data and the isotopic composition of the lead in Table 1, it is found that the U^{238}/Pb^{204} atom ratio of the glass is 38·5 and the Th^{232}/U^{238} atom ratio is 5·4. These values differ substantially from those calculated above.

Since the isotopic composition of the lead in the glass is that of a modern terrestrial lead, it must have been in an environment with "normal" ratios of uranium and thorium to lead until recent times, assuming a terrestrial origin for the present discussion. Within the last 50 to 100 million years at most uranium was enriched by a factor of 4 with respect to lead and thorium was enriched by a factor of 1·4 with respect to uranium.

The similarity of the bulk chemical composition of tektites to that of argillaceous sediments has been emphasized by many writers. The enrichment of thorium with respect to uranium in the australite further emphasizes this resemblance. The thorium would be expected to accumulate relative to uranium as a result of precipitation or adsorption processes taking place during the formation of an argillaceous sediment. Leaching processes would remove uranium preferentially to thorium. Although the arguments of this section apply only to the australite, Adams (1956) has made comparisons of the ratios of alpha activity to uranium concentration for a number of tektites and other glasses. Adams' data suggest that all tektites have high Th/U ratios similar to those found in oxidized and weathered sediments. Thus the conclusions regarding thorium–uranium differentiation probably apply to all tektites, although the magnitude of the effect can be expected to vary somewhat from one glass to another.

Suess, Hayden and Inghram (1951) have determined argon in two philippinites and an australite and set upper limits on the argon contents which correspond to maximal potassium–argon ages of from 10 to 75 million years. They believe the actual ages are probably considerably less than the quoted limits. Gerling and Yashchenko (1952) similarly set upper limits on the potassium–argon ages of a philippinite indochinite, and moldavite of from 3 to 12 million years. These are the maximum periods of time elapsed since fusion of the glasses. The uranium–thorium–lead differentiation must have preceded the fusion of the glasses, since melting alone could not be expected to produce the observed separations. For a terrestrial origin the past history of the tektites must include a chemical differentiation of elements from some source material within the last 100 million years followed by a fusion of the differentiated product within the last tens of millions of years at most. Both of these processes may actually have occurred within the last few thousands of years.

The data may be tested against possible extra-terrestrial origins for tektites. The possibilities which have been considered in the literature involve origins from meteorites, the moon or comets. The possibility of a meteoritic origin will be considered first. Simple fusion of stone meteorites is not an acceptable mode of origin. Stone meteorites with chemical compositions which correspond to those of tektites are not known, nor can a tektitic composition be produced from a stone meteorite by removal of volatile constituents. In addition tektites contain about 100 times more uranium and thorium than do stone meteorites. Thus if tektites have a meteoritic origin they must represent some special class of meteorites. The possibility that tektites are fused sediments from another planet has recently been considered by Barnes (1958). The atmospheres of all the other planets in the solar system are grossly different from that of the earth

(Kuiper, 1949). The differences make it seem unlikely that terrestrial sedimentary processes could have been duplicated on another planet.

There is moreover the problem of transporting the sediments from interplanetary space to earth in a compact swarm. Barnes uses comets for this purpose. Some evidence against comets as a source material for tektites is given below. Although it is impossible to disprove rigorously an extra-terrestrial sedimentary origin for tektites, it would seem that such a theory raises as many problems as it answers.

It appears to be difficult to reconcile the lead data with a lunar origin. Meteorites and the earth give lead results which indicate that they have evolved from matter which 4·5 billion years ago possessed lead of the same isotopic composition as that found today in Canyon Diablo and Henbury troilites. In subsequent discussion this will be referred to as the "earth–meteorite lead system." If the moon is also a member of this system, it becomes difficult to explain the uranium–thorium–lead balance for the australite. It will be recalled that the present day concentrations of uranium and thorium with respect to lead are respectively four and six times those which are required to account for the isotopic composition of the lead on the basis of the above model. The uranium–lead and thorium–lead ratios must have increased within the last few millions of years to account for the isotopic composition of the lead. It is unlikely that such a process could have taken place on the moon since it is improbable that igneous or sedimentary processes are presently taking place there. Urey (1956) recently discussed the surface features of the moon and gives evidence for believing that the craters seen on the moon were formed at a time earlier than 3 billion years ago. There are no orogenic belts on the moon such as are known on the earth, although small lunar volcanoes might exist and escape detection. Even if volcanoes are active on the moon, no terrestrial igneous processes are known which produce materials of the chemical composition of tektites so that quite different conditions would be required on the moon as compared to the earth in this case. The moon has an atmosphere of less than from 10^{-5} to 10^{-6} terrestrial atm on the hemisphere facing the sun (Struve, 1944), so that sedimentary processes must be ruled out. Alternatively, the moon may have captured large meteorites within the last tens of millions of years since it is known that the earth has done so—the Canyon Diablo meteorite, for example. The energy of these collisions would undoubtedly have produced some melted silicates such as are found near the large terrestrial meteorite craters, but it is difficult to see how this process could separate thorium from uranium in the resulting glass. The glasses found near terrestrial craters contain diagnostic quantities of nickel which are lacking in tektites (Spencer and Hey, 1933). There is, moreover, the question as to whether a group of glass particles originating from an impact on the moon could arrive at the earth in a compact swarm.

An alternative assumption is that the moon has been made in some way from matter with lead of an isotopic composition which bears no relation to that observed in the earth–meteorite lead system. Then it might be expected to be a remarkable coincidence that the lead found in tektites has an isotopic composition which closely resembles modern terrestrial lead.

Little is known about comets so that it is admittedly difficult to exclude them as a parent material for tektites. Nickel is observed in the spectra of comets along with iron, chromium and sodium when they pass near the sun (WHIPPLE, 1950). Perhaps the lack of nickel in tektites is the best evidence against their originating in comets. It might also be an unexpected coincidence if comets were to contain lead of the same isotopic composition as modern terrestrial lead.

The effect of all the arguments just discussed is to place the lead evidence concerning tektites in favour of a terrestrial origin. The difficulties encountered in an extra-terrestrial origin could be avoided by considering extra-terrestrial bodies with grossly different chemical characteristics from those of the earth–meteorite system but there is no evidence to justify such an approach at present. It is true that many questions remain unanswered once a terrestrial origin is accepted. The most serious of these is a mechanism for melting the parent materials so as to produce the observed geographic distribution of tektites. It should be noted, however, that the geographic distribution is a stumbling block to extra-terrestrial modes of origin as well. It is difficult to specify a mechanism by which tektites can arrive at the earth in a compact swarm from the moon or from interplanetary space (UREY, 1955). The problem is that if the swarm is given sufficient density in order to be gravitationally stable in the sun's gravitational field at the distance of the earth from the sun, then australites, for example, would have to be many orders of magnitude more abundant than they are.

In conclusion a general statement concerning all tektite experiments is perhaps in order. If tektites are of extra-terrestrial origin it is possible that this may be proved unambiguously by demonstrating that they have certain properties which are clearly outside the range of experience with terrestrial materials. If, on the other hand, they are of terrestrial origin, it will be impossible to prove this with certainty since our knowledge of extra-terrestrial materials is limited. The possibility will always remain that the processes which produced tektites could have been duplicated in some extra-terrestrial environment. If they are terrestrially produced objects, further experiments will only serve (in principle) to define additional areas of similarity between terrestrial materials and tektites; the greater the number of areas of similarity established, the more probable a terrestrial origin will become. This would seem to be the trend of most tektite investigations which have been conducted to date.

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Sr/Rb age study of tektites*

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Abstract—Eight tektite samples from four localities, Australia, Indo-China, the Philippines, and the Moldau Valley were analysed for Rb and Sr, and Sr isotopic composition analyses were made on seven of the samples. Seven of the eight samples are so similar in Rb/Sr ratios that a common source origin is postulated for the tektites. No radiogenic Sr^{87} was found in the tektites. All have normal 84/88, 86/88 and 87/86 ratios, namely, 0·0068, 0·1193, and 0·712. It is shown that the present Rb/Sr and Sr isotopic composition data is compatible (but unlikely) with an origin of tektites as differentiated from either proto-basaltic or achondritic material. The tektites could not have differentiated from chondritic material. Other than indicating a common and homogeneous source origin for the tektites, the present data are not evidence for or against an extra-terrestrial origin. However, the homogeneity in Rb/Sr contents and the constancy of the Sr^{87} contents make a terrestrial origin difficult to accept because of the wide geographical distributions of tektites.

Introduction

Tektites are curious objects and, as might be expected, there has been a controversy concerning their origin. Their peculiar geographical distributions and lack of resemblance to surface rocks upon which they are found scattered have suggested an extra-terrestrial origin to some investigators. They resemble obsidian in their highly vitreous character, although thin chips exhibit a greenish tint in contrast to the grey of volcanic obsidians. They are somewhat similar chemically to rhyolite, but more closely resemble in composition some high-alumina, high-silica shales.

Tektites are usually rounded, and tear-drop, pear-like, or button-like in shape. Also, their surface markings and pits and grooves are suggestive of meteoritic flight.

In a recent article Stair (1956) summarized evidence on the origin of tektites and concluded that they are extra-terrestrial. However, many investigators, ourselves included, do not believe the evidence was conclusive at the time he wrote.

Silverman (1951) measured the O^{18}/O^{16} ratio in a tektite and found it was higher than ratios found in igneous rocks, but similar to ratios found in terrestrial sediments.

Suess, Hayden and Inghram (1951) attempted to measure the ages of one australite and two phillippinites by the A^{40}/K^{40} method. The amounts of argon found were so small that measurement of A^{36}, which would presumably be atmospheric, was not possible with their equipment. Then, assuming that all the measured tektite argon was radiogenic, upper limit ages of less than, respectively 73×10^6, 10×10^6 and 32×10^6 years were calculated. However, in blank runs, comparably small amounts of argon were found. Thus the A^{40}/K^{40} ages of tektites are geologically small. Suess (1951) and his co-workers believe that diffusion of argon through

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tektites is negligible. They found the pressures in enclosed bubbles (actually voids) to be less than 10^{-3} atm and these bubbles contained no measurable argon.

Present Work

In view of the unresolved question as to the origin of tektites, we decided it would be worthwhile to measure their relative isotropic abundances of Sr to compare these abundances with various terrestrial and meteoritic materials, and to attempt to measure them by the Sr^{87}/Rb^{87} method, using the mass spectrometric isotope dilution technique. As a matter of interest we measured the K-contents flame photometrically.

Description of Samples

T3309: Tektite from French Indo-China. Given by Irving Friedman, U.S.G.S. Friedman No. 40-15. 1·6 g. Black glass chip.

T3310: Tektite from Australia. Given by Irving Friedman, U.S.G.S. Friedman No. 97-2. 2·5 g. Black glass chip.

T3311: Tektite from Philippines. Given by Irving Friedman, U.S.G.S. Friedman No. 96-2. 3·4 g. Black glass.

T3312: Tektite from Indo-China. Given by Irving Friedman, U.S.G.S. Friedman No. 96-9. 3·1 g. Black glass chip.

T3313: Tektite from Australia. Charlotte Waters, Central Australia. Given by Irving Friedman, U.S.G.S. Number not assigned to us by Friedman. 2·7 g. Black, greenish glass chip.

T3314: Tektite from Bohemia. Moldavite. Given by Irving Friedman, U.S.G.S. Number not assigned to us by Friedman. Number on the sample in white ink as follows: C. St. 20943. Pale, amber-green glass.

T3378: Tektite from Annam, Indo-China. Obtained from C. Frondel, Harvard Museum Collection.

T3379: Tektite from Philippines. Obtained from C. Frondel, Harvard Museum Collection.

Techniques

We have discussed the techniques used in this investigation elsewhere (Herzog and Pinson, 1955, 1956). The instruments used were Nier-type 60° sector, 6 in. radius mass spectrometers.

Rb^{87} and Sr^{84} spikes were added simultaneously to powdered tektite samples of known weight. The samples averaged about 0·3 g each. Separate samples of about 0·3 g each were used for the extraction of unspiked tektite Sr for isotopic ratio analyses.

The tektite Rb and Sr were extracted by ion exchange column techniques, using Dowex 50, 200–400 mesh, 8 per cent cross-linked cation exchange resin in Vycor glass columns. Platinum, Vycor and polyethylene ware was used throughout the chemical handling. Blank runs to determine contamination of Rb and Sr have repeatedly demonstrated that less than 0·2 μg of Rb and less then 0·5 μg of Sr is introduced during processing of samples.

Data were obtained by magnetic field variation sweeping over the Sr mass range, with the accelerating voltage held constant. From eighteen to eighty-three

sets of peaks were recorded for each sample. It was necessary to use three sensitivity scales (10,100 and 1000 mV) to record all peaks at heights of from 4 to 8 in. on the strip recorder. A vibrating reed electrometer was used for amplification.

The isotope ratios listed for each sample are grand averages of all peak sets measured; the errors listed (standard deviations) for the isotopic ratios are not corrected for possible isotopic fractionation of sample during analysis.

The errors assigned to the Rb and Sr analyses are based on all known sources of error, both chemical and mass spectrometric, and are believed to be real limits of error of our Rb and Sr analyses, and include such factors as uncertainties in the spike concentrations and calibrations, pipetting errors, contamination, mass spectrometric sample fractionation and peak measurement errors. We are also basing our statement of accuracy on interlaboratory comparison analyses of standard samples with the analysts at the Department of Terrestrial Magnetism in Washington, the Institute for Nuclear Studies at Chicago.

The five K-determinations of tektites were done by flame photometry, using a Beckman Model B instrument. The large errors are due to the fact that only single determinations were made, owing to the small quantities of tektite samples we had for analysis. However, these single analyses were monitored by the standard granite, G–1, a synthetic glass of "haplogranite" composition (Fairbairn, 1952) and a U.S. Bureau of Standards potassium feldspar.

Results

The tektite analytical data obtained in this investigation are presented in Tables 1, 2 and 3.

The potassium contents of $2{\cdot}3 \pm 3\%$ K compare to $1{\cdot}7 \pm 0{\cdot}2$ and $2{\cdot}2 \geq 0{\cdot}2$ reported by Suess *et al.* (1951) for two Philippinite samples, and the average value for tektites of 1·84% K given by Stair (1956).

To the best of our knowledge the only previous determinations of Rb and Sr in tektites were made by optical spectrography by Ahrens, Pinson and Kearns (1952), who reported 1·8% K, $0{\cdot}018 \pm 0{\cdot}006\%$ Rb and $0{\cdot}010 \pm 0{\cdot}003\%$ Sr in three tektites, which include two of the present samples, T3378 and T3379. The optical spectrographic analysis of Rb in the standard G–1 on which these tektite Rb analyses were based has been shown to be high by a factor of approximately 2·5 (Herzog and Pinson, 1955). On the basis of this correction factor, the revised optical spectrographic Rb analyses for the three tektites would be $0{\cdot}007 \pm 0{\cdot}002\%$ Rb, in closer agreement with the presently reported more accurate mass spectrometric tektite Rb analyses.

The K/Rb ratios for tektites reported in Table 1 average 190 with a range from 170 to 210, which is within the range of ratios found in common terrestrial rock types and chondrites, namely 220 ± 50. Only in pegmatites, certain evaporites such as sylvite, and possibly in achondrites have we found K/Rb ratios falling outside this range of values.

The strontium isotopes 84, 86 and 88 are non-radiogenic (except in fission environments). The tektite 86/88 ratios for six samples on which we have data (T3313 excluded) average 0·1193, and the standard deviations of the initial measurements all include this value. For comparison, all terrestrial and meteoritic

Table 1. Rb, K and Sr contents of tektites*

Sample no.	Locality	%K	Rb (p.p.m. by wt.)	Sr (p.p.m. by wt.)	Sr/Rb
T3309	Indo-China	2·2 ± 0·3	126 ± 6	117 ± 6	0·9
T3312	Indo-China	2·1 ± 0·3	110 ± 6	111 ± 6	1·0
T3378	Indo-China	—	110 ± 6	120 ± 6	1·1
T3313	Australia	2·1 ± 0·3	111 ± 6	141 ± 7	1·3
T3310	Australia	2·1 ± 0·3	110 ± 6	284 ± 15 298 ± 15	2·6
T3311	Philippines	2·4 ± 0·4	141 ± 7	150 ± 7	1·1
T3379	Philippines	—	104 ± 6 112 ± 6	162 ± 8	1·5
T3314	Bohemia	2·7 ± 0·4	130 ± 7	136 ± 7	1·0

* K by flame spectrometry.
* Rb and Sr by mass spectrometric stable isotope dilution.

Table 2. Sr isotopic composition of tektites

Sample no.	Locality	84/88	86/88	87/86	% Corr. to 87/86	No. sets
T3309	Indo-China	0·0068 ± 0·0001	0·1199 ± 0·0005	0·712 ± 0·002	−1·1	83
T3312	Indo-China	0·0068 ± 0·0001	0·1199 ± 0·0005	0·712 ± 0·002	−1·4	60
T3378	Indo-China	0·0069 ± 0·0001	0·1185 ± 0·0005	0·715 ± 0·004	−1·5	18
T3313	Australia	0·0066 ± 0·0003	0·1185 ± 0·0010	0·695 ± 0·008	−5·4	30
T3310	Australia	0·0069 ± 0·0001	0·1200 ± 0·0005	0·709 ± 0·002	−1·3	80
T3311	Philippines	0·0067 ± 0·0002	0·1192 ± 0·0005	0·712 ± 0·002	−1·0	32
T3379	Philippines	0·0072 ± 0·0004	0·1191 ± 0·0006	0·712 ± 0·003	−2·1	40

Tertiary basalts (P. Gast, private communication) Rb/Sr ratio ≅ 0·1	0·708
Sea water (Pinson *et al.*, 1957b)	0·712
Normal Sr (Bainbridge and Nier, 1950)	0·712

Sr samples analyzed thus far average about 0·1195 with a range from about 0·1185 to 0·1205, part of which variation may well be due to analytical and instrumental errors of measurement. The tektite is not unusual in this respect. The 84/88 ratio is similarly normal. For samples T3312, T3310 and T3309 the standard deviations reported for the 84/88 ratios are the smallest we have achieved thus far. This high precision was due to unusually strong and steady Sr emission throughout the runs.

Isotopic ratios of the unspiked Sr extracted from the seven tektites are reported in Table 2.

The ratio 87/86 in tektites is as normal as it can be. If we had analysed the U.S. Bureau of Standards Sr seven times under identical conditions (and the runs have had about the same quality range as the present analyses do), we well might have obtained a set of values very similar to the 87/86 ratios reported in Table 2 for the seven tektites. In illustration, we have included the average of two analyses of U.S. Bureau of Standards Sr sample No. 12. Sr isotope ratio analyses for the

Table 3. Isotopic composition of Sr from various source materials

Sample	Source	84/88	86/88	87/86	Sr^{87}/Sr	Sr/Rb
Tektites average T3313 excluded	Present work	0·0068 ± 0·0001	0·1193 ± 0·0003	0·712 ± 0·002	—	1·1
Young basalts	GAST (pvt. comm.)	—	—	0·708	—	~10
Normal Sr	BAINBRIDGE and NIER, 1950	0·0068	0·1194	0·712	0·0702	—
Normal Sr	ALDRICH and HERZOG, 1953	0·0067 ±0·0001	0·1195 ± 0·0003	0·711 ± 0·003	0·0702	—
Normal Sr (1st analysis)	MIT, NBS, $SrCO_3$ Sample No. 12	0·0068 ± 0·0002	0·1196 ± 0·0003	0·712 ± 0·002	0·0703	—
Normal Sr (2nd analysis)	MIT, NBS, $SrCO_3$ Sample No. 12	0·0068 0·0002	0·1193 0·0003	0·713 0·002	0·0703	
Sea water (mid-Atlantic)	PINSON *et al.*, 1957b	0·0067 ± 0·0001	0·1189 ± 0·0003	0·712 ± 0·002	0·0702	62
Grenville limestone Sr	MIT, unpublished data	0·0068 ± 0·0002	0·1195 ± 0·0003	0·711 ± 0·002	0·0702	>1000
Homestead chondrite	HERZOG and PINSON, 1956	0·0069 ± 0·0002	0·1183 ± 0·0005	0·753 ± 0·004	0·0734 ± 0·0002	3·0 ± 0·2
Forest City chondrite	HERZOG and PINSON, 1956	—	0·1194 ± 0·0005	0·755 ± 0·004	0·0736 ± 0·0004	2·8 ± 0·2
Forest City chondrite	SCHUMACHER, 1956 a, b	"normal"	"normal"	—	0·0735	3·1
Pasamonte achondrite	HERZOG and PINSON, 1956	0·0068 ± 0·0003	0·1197 ± 0·0008	0·697 ± 0·009	0·0689 ± 0·0009	80
Pasamonte achondrite	SCHUMACHER, 1956 a, b	"normal"	"normal"	—	0·0676 ± 0·0003	170

moldavite T3314 were all failures, and the analysis of the australite T3313 was of such poor quality that we hesitated to report it.

The isotope ratios reported in Table 2 are different from those orally reported by us at the June 1957 Conference on Tektites held at the National Academy of Sciences in Washington and at the November 1957 Annual Meeting of the Geological Society of America, and printed in the Program Abstracts of that meeting. *An examination of these mass spectrometric records during preparation of this paper for publication revealed that corrections to the recorded peak heights of masses* 84,86 *and* 87, *necessary when the much larger* 88 *peak "tails off" due to "pressure broadening" and to other instrumental factors, had not been made. Responsibility for this blunder is entirely that of the senior author.* The magnitudes of these corrections to the isotope ratios were small, ranging from 1 to 2 per cent for the ratio 87/86, and were smaller for the other ratios, but are sufficient to *invalidate the conclusions* then reported that the tektites contained measurable amounts of radiogenic Sr^{87}, as previously reported.

As reported in Table 2, obviously, if an initial 87/86 ratio of 0·712 is assumed, the tektite samples have developed no radiogenic increments of radiogenic Sr^{87} and must be very young, too young to be dated by the Sr^{87}/Rb^{87} method. Of course, the analytical errors that must be assigned to the 87/86 ratio measurements permit "ages" of up to about 75×10^6 years, but even more strongly indicate ages of essentially zero on a geological time scale.

These revised results are therefore in complete agreement with the A^{40}/K^{40} method analyses for ages reported by SUESS *et al.* (1951), namely less than 75×10^6 years. Neither radiogenic Sr^{87} nor radiogenic A^{40} has been found in these objects.

Discussion of Results

One is of course permitted to differ with our choice of an initial 87/86 ratio of 0·712. Since we do not know the source material from which tektites were derived it is speculative and arbitrary to select a value. It may, however, be of interest to use the present data to test various hypothetical source materials for the tektites, e.g. terrestrial basalts, chondritic and achondritic meteorites, a "shattered planet,"

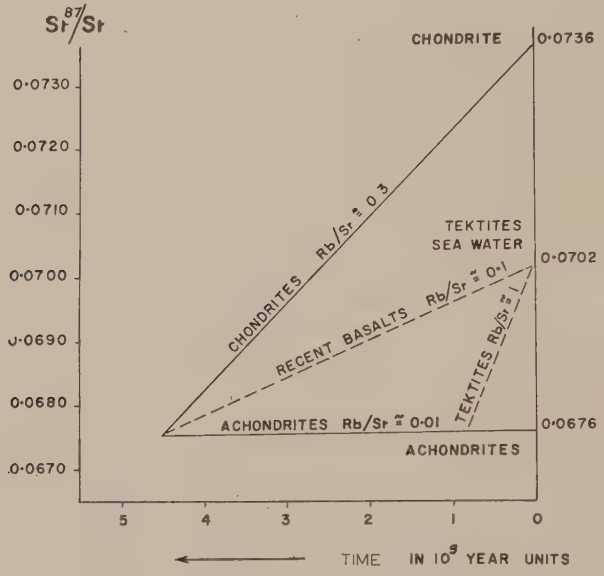

Fig. 1. Variation of Sr^{87}/Sr with time for meteorites, basalt and tektites.

and moon surface material. For this purpose several analyses of isotopic ratios of Sr from various materials have been compiled in Table 3, and some of these data have been plotted in Fig. 1.

(1) *Terrestrial basalts.* Dr. Paul Gast of Lamont Geological Observatory has made the only isotope ratio Sr measurements to date on recent basalts. He has communicated to us an average value for 87/86 in the samples of 0·708. The limits of error of our tektite values and his basalt Sr values overlap. Thus the tektite data are compatible with an origin of tektites from proto-basalts as differentiates (they would have to be differentiates because their compositions are considerably different from basalts) in Recent or Tertiary times.

(2) *Terrestrial surface rocks.* The bulk chemical composition of tektites is similar to that of some high-silica, high-alumina shales, and it is conceivable that meteoritic impact into such a terrestrial rock could have produced the glassy tektites ("impactites"). We have no information on the isotopic composition of Sr from shales and therefore cannot compare the 87/86 ratios found in tektites to shale Sr. However, since most shales are sedimentary derivatives of pre-existing rocks, some of which (as for example, granites) probably have 87/86 ratios always greater than 0·712, it is probably not possible that tektites were derived from shales or other

detrital sedimentary derivatives of pre-existing terrestrial rocks. Furthermore, the constancy of the 87/86 ratio in tektites strongly negates the possibility of their being melted derivatives of any pre-existing acidic rocks. Remelting of basalt is excluded because of the greatly different bulk chemical composition of basalt, but the very slim possibility of an impact derivation from rhyolite remains. However, the $Sr^{87}/86$ ratios in rhyolites are unknown, and the relative scarcity of outcrops of rhyolite on the earth's surface makes the impact hypothesis for such a rock highly improbable.

Some recent marine sediments in shallow waters, in which most of the Sr may have been precipitated from sea water, remain as a possible tektite source, however, both on the basis of their probable lower 87/86 Sr ratios and their greater constancy in chemical composition.

(3) *Chondritic meteorites.* We have analysed two chondrites (Herzog and Pinson, 1956), one of which was also analyzed by Schumacher (1956 a,b). All results for present day Sr^{87}/Sr from chondrites are in the range $0{\cdot}0735 \pm 0{\cdot}002$. Since the Rb/Sr ratio in these samples is approximately $\frac{1}{3}$, while the Rb/Sr is approximately 1 in tektites, it can be seen from Fig. 1 that tektites cannot have been formed by differentiation of chondritic material. Furthermore, because of the demonstrated constancy of chemical composition of chondrites (notably here in their Rb and Sr contents) as reported by Ahrens *et al.*(1952), and Edwards and Urey (1955), we believe these isotopic Sr ratios on two chondrites, Homestead and Forest City, are representative of chondrites as a group.

(4) *Achondritic meteorites.* These meteorites have a wide range of chemical compositions, but the only ones thus far analysed for Rb, Sr and Sr isotopic composition were chosen because they had extremely-large Sr/Rb ratios (about from 80 to 200); so large, in fact, that only a negligible increase in Sr^{87} relative abundance could have occurred in them from 5×10^9 years of Rb^{87} decay. Schumacher (1956) found that two achondrites had present Sr^{87}/Sr (fraction of Sr^{87} divided by total Sr) of 0·0675 and 0·0679, that is 87/86 ratios of 0·685 and 0·689, in agreement with the lower limit of our own measurement of the same quantity (Herzog and Pinson, 1956).

Cassidy (article, this Symposium on Tektites, pp. 304–315) has suggested that tektites and achondrites may be cogenetic or that tektites may have been derived from achondritic material. A contemporaneous tektite–achondrite origin is excluded if, as shown in Fig. 1, it is assumed that the chondrites and achondrites formed contemporaneously, approximately $4{\cdot}5 \times 10^9$ years ago. Both Schumacher, and Herzog and Pinson made this assumption of contemporaneous origin in dating the meteorites by the Sr^{87}/Rb^{87} method. The slopes of the lines are of course determined by the Rb/Sr ratios in the meteorites, with a known point at the present day Sr^{87}/Sr ratio. The chondrite and achondrite lines intercept at about $4{\cdot}5 \times 10^9$ years ago. However, the slope of the tektite lines, having an average Rb/Sr ratio of about 1 and a present Sr^{87}/Sr ratio of 0·0702 is much greater than the chondrite line. It intersects the achondrite line approximately 800×10^6 years ago, but does not intersect the chondrite line; thus the tektites could not have differentiated from chondritic material. Had the tektites differentiated $4{\cdot}5 \times 10^9$

years ago from chondritic material the present day Sr^{87}/Sr ratio should be about 0·0825. The observed value is $0·0702 \pm 0·0003$.

The intersections of the tektite and achondrite lines at 800×10^6 years makes possible an achondrite source origin for the tektites. However, we believe that the fact that this fictitious date is greatly different from $4·5 \times 10^9$ years excludes such an origin—unless a valid mechanism for remelting, strong differentiation, and solidification of the achondritic material at 800×10^6 years ago can be discovered.

(5) A "*Shattered planet.*" If all meteorites were formed as part of the breakup of a planet, similar in composition and structure to the theoretical model of the earth, the Rb/Sr ratios and the 87/86 Sr ratios for the tektites indicate a very recent breakup. However, studies of meteorites indicate that they have been exposed to outer space cosmic-ray flux for very long periods. For example, Fireman and Schwarzer (1957) found a minimum cosmic-ray age of $0·9 \times 10^9$ years for the Norfolk (iron) meteorite, $1·7 \times 10^9$ years for the Para de Minas (iron) meteorite, indicating a hypothetical planetoid breakup occurring not less than about 10^9 years ago. These "cosmic-ray ages" were based on studies of the Li^6, He^3 and H^3 contents of iron meteorites. Therefore, the very recent Sr^{87}/Rb^{87} and A^{40}/K^{40} tektite ages seem to exclude the possibility that tektites are fragments of a shattered planet.

(6) *Splashes from the moon.* Varsavsky (1957) (and also in this Symposium on Tektites) has theorized that tektites may be lunar material "splashed off" when large meteorites collide with the moon, and material is focused by the earth-moon gravitational system. If so, our measurements indicate that lunar surface material is very similar to average terrestrial surface material (e.g. basalt or sea water Sr) in Sr isotopic composition. However, the tektite Sr isotopic and Rb/Sr data shed *per se* no light on a possible lunar origin of tektites.

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Cosmic-ray-induced radioactivities in meteorites—I

Chemical and radiometric procedures for aluminium, beryllium and cobalt*

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Abstract—It is expected that the long-lived radionuclides Al^{26}, Be^{10} and Fe^{60} (with its daughter Co^{60}), among others, are present in detectable amounts in meteorites as the result of the action of cosmic rays in space. For the detection and measurement of these activities, which have considerable potential significance, chemical isolation and purification are essential.

Chemical procedures developed for the separation and purification of aluminium, beryllium, iron and cobalt from iron and stone meteorites, tektites, and terrestrial silicate materials are described in detail. The prime objective was radiochemical purity of the isolated radioelements, chemical purity and chemical yields being of secondary importance. Ion-exchange and solvent-extraction techniques were used wherever possible because of their high specificity and applicability to very small quantities of elements.

Al^{26} and Be^{10} are prepared for counting in the forms of Al_2O_3 and BeO. Fe^{60} is best detected by extraction of its active daughter Co^{60}, counted as CoS. The identity of a radionuclide is established by the criterion of constant specific activity upon recycling the sample through the chemical purification procedure.

Special low-background counters have been constructed for the detection and measurement of feeble amounts of beta emitters in the form of small solid samples. The backgrounds are about 0·4 count min^{-1} and are exceptionally constant. In routine operation 3–4 day backgrounds are alternated with 6–8 day sample counts, yielding a lower counting rate limit (2σ criterion) of 0·017 count min^{-1}. With counting yields of about 40 per cent and specimen sizes of about 100 g, it is possible to detect radionuclides at a specific activity of 0·0004 dis min^{-1} (g specimen)$^{-1}$ in favourable cases.

Thin-sample counting yields have been measured for Al^{26} and Na^{22}. Sample-thickness factors have been determined for Al^{26} in Al_2O_3 and Be^{10} in BeO.

1. Introduction

This and the following paper describe the techniques, apparatus, and results of some experiments undertaken to detect and measure long-lived cosmic-ray-induced radioactivities in meteorites. The possibility that detectable amounts of radionuclides should result from nuclear reactions induced in meteorites exposed to the free-space cosmic-ray flux was first suggested by Bauer (1947), and was strengthened by the detection of similarly produced stable isotopes of helium, neon and argon. The first radioactive product to be detected was tritium (Fireman and Schwarzer, 1957; Begemann, Geiss and Hess, 1957).

Studies of such radioactivities may be of significance for (1) dating falls of meteoritic "finds", (2) dating times of break-up of larger bodies into meteoroids, (3) calibrating similar dating methods based on stable reaction products, (4) estimating sizes and shapes of pre-atmospheric meteoroids and amounts of material ablated from them, (5) testing for cosmic origin of objects of suspected meteoritic nature (especially tektites), and (6) providing information about the past intensity of the cosmic radiation. These possibilities have been discussed elsewhere (Kohman and Ehmann, 1958), and will be dealt with again in the second paper of this series (Ehmann and Kohman, 1958).

We have been particularly interested in very long-lived radioactive products, so as to be able to extend studies of the above-mentioned kinds as far back into

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the past as possible. The abundances in the cosmic radiation of particles having atomic number $Z > 2$ and in meteorites of target atoms having $Z > 28$ are so low as to preclude the production in appreciable amounts of nuclides having $Z > 30$. Table 1 lists with their disintegration characteristics all known nuclides

Table 1. Possible cosmic-ray-induced radionuclides in meteorites

Nuclide	Half-life (yr)	Reference	Radiations	Daughter and radiations
Be^{10}	$2{\cdot}7 \times 10^6$	(a, b)	β^-	
Al^{26}	8×10^5	(c)	$\beta^+, \epsilon, \gamma$	
Cl^{36}	$3{\cdot}08 \pm 0{\cdot}03 \times 10^5$	(d)	β^-, ϵ	
Fe^{60}	$\sim 3 \times 10^5$	(e)	$\beta^-, \gamma(?)$	Co^{60m};Co^{60g} I.T.; β^-, γ
Mn^{53}	$\geq 1{\cdot}3 \times 10^5$	(f)	ϵ	
Ca^{41}	$1{\cdot}1 \pm 0{\cdot} \times 10^5$	(g)	ϵ	
Ni^{59}	$1{\cdot}0 \pm 0{\cdot}2 \times 10^5$	(h)	ϵ	
C^{14}	$5{\cdot}67 \pm 0{\cdot}03 \times 10^3$	(i)	β^-	
Ti^{44}	$\sim 1 \times 10^3$	(j)	ϵ, γ	Sc^{44} $\beta^+, \epsilon, \gamma$
Si^{32}	$\sim 7 \times 10^2$	(k)	β^-	P^{32} β^-
A^{39}	265 ± 30	(l)	β^-	
Ni^{63}	125 ± 6	(m)	β^-	
H^3	$12{\cdot}262 \pm 0{\cdot}004$	(n)	β^-	
Co^{60}	$5{\cdot}20 \pm 0{\cdot}03$	(o)	β^-, γ	
A^{42}	$\geq 3{\cdot}5$	(p)	β^-	K^{42} β^-, γ
Fe^{55}	$2{\cdot}60 \pm 0{\cdot}02$	(q)	ϵ	
Na^{22}	$2{\cdot}58 \pm 0{\cdot}03$	(r)	$\beta^+, \epsilon, \gamma$	

ϵ indicates electron capture, with X-radiation and Auger electrons.
I.T. indicates isomeric transition, with γ radiation, conversion electrons, X-radiation, and Auger electrons.

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with $Z \leq 30$ and half-life between 1 year and 10^9 years. Fig. 1 shows the distribution on a logarithmic scale of the half-lives of these nuclides. Shorter-lived nuclides would also be of interest, but present special problems in connexion with the difficulties of obtaining freshly-fallen materials and the necessity for rapid operations.

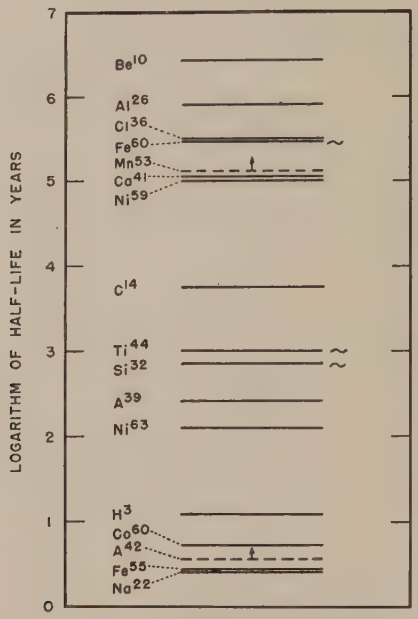

Fig. 1. Possible cosmic-ray-induced radionuclides in meteorites with half-lives greater than 1 year.

2. Chemical Procedures

A. Objectives and General Nature

Chemical separation and purification of individual radioelements is essential for concentration of activities for measurement, for elimination of disturbing primary and secondary natural radioactivities, and for positive identification of any activities detected. Our chemical procedures were devised to yield, in addition to Be^{10} and Al^{26}, the two longest-lived nuclides listed in Table 1, Ca^{41}, $Fe^{55,59,60}$, Co^{60} and $Ni^{59,63}$. Cl^{36} was not included because of its volatility under conditions used for dissolving meteorites, and Mn^{53} was not included because until recently it was believed to be relatively short-lived. Co^{60} was included because it can serve as an indicator for its long-lived parent Fe^{60} (Roy and Kohman, 1957).

Only the procedures for Be, Al and Co have been completely tested so far by counting of the isolated elements and are reported here. Chemical techniques for the specific purification of Ni and Ca while not discussed in detail here may be found elsewhere (Ehmann, 1957).

The objectives of the procedures were, in order of importance, high radiochemical purity of each isolated radioelement, high chemical purity of the sample, and reasonably high chemical yield.

The procedures are of three types: (1) for iron meteorites, (2) for stone meteorites, tektites, and other silicate materials, and (3) for recycling. By "recycling" is meant the dissolution and further purification of an isolated sample for the purpose of establishing the identity, purity, and activity level of the particular radionuclide. Only if the specific activity of a sample remains constant after appropriate recycling can the presence, identity, purity, and activity level be regarded as definitely established. The activity levels are generally too low to permit radiation characterization as a reliable means of certification.

The compositions of various materials to which the procedures have been applied are listed in Tables 2 and 3. In one or two instances the published cobalt

Table 2. Composition of the silicate phase of chondrites, tektites and terrestrial silicates

Material (wt. fr.)*	Plainview (84·21%) ‡	Richardton (80·10%)	Australites (100%)	Bediasites (100%)	Moldavites (100%)	Libyan D. silica-glass (100%)	Granite (typical) (100%)	Obsidian (typical) (100%)
†	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)
SiO_2	41·45	44·49	70·62	77·76	74·91	98·20	72·55	74·7
Fe_2O_3	3·46	4·34	0·85	0·37	0·44	0·53	1·41	1·01
FeO	17·77	16·23	4·44	3·36	3·10	0·24	0·54	0·62
Al_2O_3	3·17	2·46	13·48	13·30	13·80	0·70	14·29	13·72
Cr_2O_3	0·40	0·64	—	—	—	—	—	—
Na_2O	0·90	0·85	1·27	1·41	0·72	0·33	4·92	3·9
K_2O	0·14	0·165	2·22	1·97	3·76	0·02	5·06	4·02
CaO	1·63	2·69	3·09	0·04	1·16	0·30	0·17	0·78
MgO	28·99	28·32	2·42	1·19	1·66	0·01	0·06	0·14
MnO	0·21	0·19	0·42	0·01	0·02	—	0·02	—
TiO_2	—	0·10	0·90	0·76	0·55	0·23	0·25	—
CoO	0·04	—§	—	—	—	—	—	—
NiO	0·91	—	—	—	—	0·02	—	—
H_2O	—	—	0·07	0·02	0·12	0·06	0·23	0·62
Reference	(a)	(b)	(c)	(c)	(d)	(d)	(e)	(f)

References for Table 2

* The numbers in parentheses are the weight fractions of silicate phase in the indicated materials.

† The other entries in the table are the weight fractions of the indicated constituents in the silicate phase of the indicated materials.

‡ Troilite (FeS) is listed separately as 9·57%.

§ Our analysis: 0·070% Co in gross material of Richardton chondrite. In connexion with the Co content of the metal phase (Table 3), this would correspond to 0·050% Co in the silicate phase.

(a) Merrill G. P. (1917) A new find of meteoritic stones near Plainview, Hale County, Texas. *Proc. U.S. Nat. Mus.* **52,** 419.

(b) Quirke T. T. (1919) The Richardton meteorite. *J. Geol.* **27,** 431.

(c) Barnes V. E. (1940) *North American Tektites.* University of Texas Publ. No. 3945, 477.

(d) Spencer L. J. (1939) Tektites and silica-glass. *Min. Mag.* **25,** 425.

(e) Tilton G. R., Patterson C., Brown H., Inghram M., Hayden R., Hess D. and Larsen E., Jr. (1955) Isotopic composition and distribution of lead, uranium, and thorium in a precambrian granite. *Bull. Geol. Soc. Amer.* **66,** 1131.

(f) Hubbard D., Krumrine E. M. and Stair R. (1956) Australite (meteoritic) glass. *Trans. Amer. Geophys. Un.* **37,** 767.

analyses appear to be in error; in each case the cobalt content obtained by our analysis is given below the table. The rest of the data in the tables appears to be consistent with the yields we obtained in those instances where our procedures are essentially quantitative.

Table 3. Composition of siderites and metal phase of chondrites

Material (wt. fr.)*	Canyon Diablo (~99%)	Canyon Diablo spheroids (>50%)	Canyon Diablo spheroids (metallic centres)	Odessa (~99%)	Plain-view (6·22%)	Richardton (19·90%)
†	(%)	(%)	(%)	(%)	(%)	(%)
Fe	90·43	53·88	75·63	90·30	90·63	90·89
Co	0·49	0·95	2·22	0·15‡	0·38	0·15
Ni	8·54	9·06	17·30	8·73	8·85	8·92
P	0·34	0·56	—	0·81	0·14	0·04
SiO_2	0·08	—	—	—	—	—
Cr	0·02	—	—	trace	—	—
Cu	0·04	—	—	—	—	—
S	0·13	0·96	—	0·02	—	—
CaO	—	1·10	—	—	—	—
Insol. res.	—	16·10	1·15	—	—	—
Reference	(a)	(b)	(b)	(c)	(d)	(e)

References for Table 3

* The numbers in parentheses are the weight fractions of metal phase in the indicated materials.
† The other entries in the table are the weight fractions of the indicated constituents in the metal phase of the indicated materials.
‡ Our analysis: 0·36% Co in Odessa siderite.

(a) Buddhue J. D. (1950) New chemical analyses of the Canyon Diablo, Arizona, and Arispe Sonora, Mexico, siderites. *Pop. Astr.* **58,** 190.
(b) Nininger H. H. (1952) *Out of the Sky*, p. 242. University of Denver Press, Denver, Colorado; (1956) *Arizona's Meteorite Crater*, pp. 88 and 106. World Press, Denver, Colorado.
(c) Beck C. W. and LaPaz L. (1951) The Odessa, Texas, siderite, ECN = +1025, 318. *Pop. Astr.* **59,** 145.
(d) Merrill G. P. (1917) A new find of meteoritic stones near Plainview, Hale County, Texas. *Proc. U.S. Nat. Mus.* **52,** 419.
(e) Quirke T. T. (1919) The Richardton meteorite. *J. Geol.* **27,** 431.

The overall scheme of these separations is original, but many of the individual steps are modifications of procedures previously published. Ion-exchange and solvent-extraction techniques are used wherever possible because of their high specificity and applicability to small quantities of elements. Some of the separations which make use of anion exchange were derived from the excellent work of Kraus and Nelson (1956). The $AlCl_3$ precipitation step for aluminium (Hillebrand and Lundell, 1953) and the solvent extraction steps for iron (Gilmore, 1953) and cobalt (Burgus, 1953) were adapted for our specific purposes from previously published work. The purification scheme for beryllium is original and gives a chemical yield higher than other radiochemical separation schemes for this element described in the literature, including that of Arnold (1956).

The procedures should be essentially specific for each element of interest with respect to all other elements of $Z \leq 30$ and all primary and secondary natural radio-elements. The iron may be accompanied by trace amounts of Ga, As, Mo, In, Sb, Te, Au and Tl, since these also extract into ether from hydrochloric acid solutions; but these are minor constituents of meteorites and are not naturally radioactive (except for the very feeble activity of In^{115}, which is well below the limit of detection). However, as will be seen, the beryllium samples often required several recycles to reduce their activity to a constant level or to zero, and there was evidence that the separation of some macroscopic impurity from this element was also incomplete without moderate recycling.

The procedures are given in detail for iron meteorites. For silicates the separation scheme is quite similar, but differs in the method of sample dissolution and in amounts of reagents and sizes of ion-exchange columns necessitated by the relatively large amounts of magnesium and aluminium. The recycle procedures are mainly adaptations of steps from the initial separation and purification procedures. Accordingly, the procedures for silicate materials and the recycle procedures refer frequently to the procedures for iron meteorites.

B. *Materials*

Reagent chemicals. Ordinary reagent-grade chemicals are used throughout these procedures. Standard carrier solutions for this work were also prepared with reagent chemicals. The NH_4OH used is fresh reagent-grade containing less than 0·002 per cent CO_2. All water is freshly distilled water from the laboratory still.

Filter media. Ordinary analytical grades of Whatman filter paper were used in most filtrations performed in this work.

In a few instances, Millipore HA filter papers, obtained from the Millipore Filter Corporation, Watertown 72, Massachusetts, were used. These papers have a very small pore size and were particularly useful in the quantitative filtration of small amounts of finely divided $Al(OH)_3$ and $Be(OH)_2$. Upon ignition these papers are ashless.

Ion-exchange resins. The ion-exchange resins used in this work were manufactured by the Dow Chemical Company, Midland, Michigan. "Dowex-1", as used in these procedures, is Dowex-1, X-10, 100–200-mesh low-porosity anion-exchange resin. "Dowex-50", as used in these procedures, is Dowex-50, X-8, 50–100-mesh medium-porosity cation-exchange resin.

In each of the ion exchange steps in these procedures, the resin is rinsed in a beaker with distilled water several times and the "fines" are decanted off. The resin is then introduced into a glass burette of the appropriate size. The resin is supported in the burette on a small wad of glass wool. The column of resin is rinsed with ten resin volumes of the eluting solution to be used in a given separation before the introduction of the sample.

pH *Indicators.* In those steps where only an approximate pH determination is required Alkacid Test Paper supplied by Fisher Scientific Company, is used. Where a more accurate pH determination is required, methyl red indicator or a Beckman Model H2 pH meter is used as indicated in the procedures.

C. *Procedure for Iron Meteorites*

(a) *Sample preparation and solution*

(1) An individual sample of the iron meteorite weighing from 100 to 150 g is washed with distilled water and acetone to remove terrestrial dirt and any lacquers which may have been used to preserve the specimen. If small complete individual pieces of the meteorite are not available, samples are cut from larger specimens using a power-driven hacksaw. Samples having traces of a fusion crust are selected where possible, in order that shielding from the cosmic rays may be minimized.

(2) The sample is placed in a 2 l. beaker and treated with consecutive 200 ml portions of aqua regia. After reaction has ceased, each portion is poured off into a separate beaker. This treatment is continued until the specimen is completely dissolved.

(3) Approximately 25 ml of concentrated HNO_3 is added to the combined solutions to assure oxidation of all Fe^{+2} to Fe^{+3}, and the solution is evaporated to near dryness with several 500 ml portions of concentrated HCl to remove the excess HNO_3.

(4) The small volume of solution from the last evaporation is made up to approximately 1 l. with 9 M HCl. This solution is filtered, using Whatman-50 filter paper, in a Büchner funnel. The small amount of residue on the filter (possibly graphite) is discarded.

(5) Carriers for Al^{+3} (20 mg), Be^{+2} (13·9 mg) and Ca^{+2} (20 mg) are added to the 9 M HCl solution of the sample. The quantities added must be known accurately, to permit chemical yield determinations.

(b) *Iron extraction*

(6) The solution is solvent-extracted in a 2 l. separatory funnel with consecutive 300–400 ml portions of 9 M HCl-saturated *iso*propyl ether. The aqueous layer is drawn off the bottom into a clean separatory funnel and the organic layer is poured out the top into a beaker after each extraction. The Fe^{+3} is usually quantitatively extracted into the ether layers after three or four extractions.

(7) The ether layers containing the Fe^{+3} are combined and washed three times with 50 ml portions of 9 M HCl. These washings are added to the aqueous phase, which is further processed in step 8.

The iron in the ether phase is back-extracted with 3 or 4 100 ml portions of distilled water. The iron may be electrodeposited on to a copper sheet for direct counting of $Fe^{55,59,60}$ in a large internal sample counter, or it may be held in solution for Co^{60} growth and subsequent isolation.

(c) *General anion-exchange separation*

(8) The aqueous phase remaining from the iron extraction is reduced in volume to approximately 250 ml on the hot plate. To this solution is added 500 ml of 12 M HCl, making the resultant solution approximately 10 M in HCl.

This solution is passed through an ion exchange column, approximately 2·5 cm. in diameter, containing 200–250 ml wet volume of Dowex-1. The flow rate is adjusted to approximately 1 ml/min. Residual Fe^{+3}, Co^{+2}, Cr^{+6}, U^{+4}, U^{+6}, Pa^{+5}, Po^{+4}, Bi^{+3}, and about thirty other elements are held in the column ($\log D \geq 1$); while Ni^{+2}, Al^{+3}, Ca^{+2}, Th^{+4}, Pb^{+2}, Ra^{+2} and Ac^{+3} of the elements of interest pass freely through (no adsorption). The alkali elements and the other alkaline earth elements are also not absorbed. The column is washed with 500 ml of 10 M HCl, and the eluates combined and saved for step 13.

(d) *Cobalt purification*

(9) Co^{+2} is eluted from the column with approximately 400 ml of 4 M HCl, and the resulting solution containing Co^{+2} is reduced in volume to 25 ml on the hot plate.

Most of the elements still adsorbed on the column are eluted with five resin volumes of distilled water. These may be saved for future reference, if desired.

(10) The cobalt-containing solution of step 9 is diluted to 250 ml with distilled water. To this solution are added 50 g of NH_4SCN, forming the blue cobalt thiocyanate complex. Sodium citrate (1 g) is added to prevent formation of the red iron thiocyanate complex, which would also extract if traces of iron are still present. The solution should be slightly acidic at this point.

The cobalt thiocyanate complex is extracted into 100 ml portions of 1 : 1 amyl alcohol–ethyl ether in a 500 ml separatory funnel. The aqueous phase is drawn off the bottom of the separatory funnel into a clean separatory funnel. The organic layer is poured out of the top of the funnel into a beaker. The extraction is repeated with additional portions of the organic mixture. Usually, from 3 to 5 extractions are required to remove all the blue cobalt complex from the aqueous phase.

(11) The organic layers are combined and washed three times with 25 ml portions of a

saturated aqueous solution of NH_4SCN in a separatory funnel. The aqueous layer and the washings are discarded.

The cobalt is back-extracted into successive 100 ml portions of 6 M NH_4OH until no further colour is removed from the organic layer. Usually, three or four extractions are sufficient.

(12) The aqueous cobalt solution is heated to expel dissolved ether and excess NH_3. CoS is precipitated from the hot, slightly alkaline solution with H_2S gas. The precipitate is filtered on Whatman-50 filter paper in a 6·5 cm Büchner funnel, using low suction, and washed with distilled water.

The CoS is ignited in a platinum dish at 800° for $\frac{1}{2}$ hr in a muffle furnace. The CoS is then mounted on an aluminium dish for counting.

(e) *Separation of aluminium and beryllium from calcium, nickel, and other elements*

(13) The solution containing Ni^{+2}, Be^{+2}, Al^{+3} and Ca^{+2} from step 8 is reduced in volume to 300 ml on the hot plate. $Al(OH)_3$ and $Be(OH)_2$ are precipitated by the addition of NH_4OH to pH ~7. Most of the nickel, together with calcium, magnesium, and the other alkaline earths, remain in solution. The insoluble hydroxides are filtered on Millipore HA filter paper in a 6·5 cm Büchner funnel and washed with 25 ml 5% NH_4Cl adjusted to pH ~7.

$Al(OH)_3$ and $Be(OH)_2$ (containing traces of nickel) are dissolved in dilute HCl and reprecipitated and filtered twice as above. These filtrates are added to the previous filtrate and saved for step 16.

(14) The $Al(OH)_3$ and $Be(OH)_2$ are transferred on the filter paper to a 250 ml beaker and are treated with 25 ml of 8 M NaOH and 10–20 mg of Fe^{+3} carrier. The paper is macerated and the mixture is heated to boiling. The warm slurry is filtered through a funnel having a 6·5 cm fritted glass disk. $Be(OH)_2$ and $Al(OH)_3$, being amphoteric, dissolve in the NaOH and pass through the filter, while residual traces of Ni, Mn, Ti, Th and other impurities form insoluble hydroxides which are carried down by $Fe(OH)_3$ and are filtered out. The precipitates are dissolved in dilute HCl and saved for step 16, or discarded if their Ni content is small.

(15) The filtrate containing Al^{+3} and Be^{+2} is diluted to 250 ml with distilled water and treated with 6 M HCl to reprecipitate $Al(OH)_3$ and $Be(OH)_2$ at the methyl red end-point. These hydroxides are filtered, dissolved in dilute HCl, and reprecipitated twice with NH_4OH at the methyl red end-point to remove most of the Na^+, and saved for step 19.

(f) *Separation of nickel from calcium*

(16) The filtrates from step 13 containing mainly Ni^{+2} and Ca^{+2} and any nickel-containing solution from step 14 are combined and reduced in volume to 500 ml. NiS is precipitated from the hot, slightly alkaline solution with H_2S gas. The solution is then boiled for about 10 min to remove the excess H_2S. The NiS is filtered on Whatman-50 filter paper, in a 12 cm Büchner funnel, washed with distilled water and saved for step 18.

(g) *Specific purification of calcium*

(17) Calcium may be further purified using methods discussed elsewhere (Ehmann, 1957).

(h) *Specific purification of nickel*

(18) Nickel may be further purified using methods discussed elsewhere (Ehmann, 1957).

(i) *Separation of aluminium from beryllium*

(19) The $Be(OH)_2$ and $Al(OH)_3$ from step 15 are dissolved in the minimum amount of 6 M HCl necessary to yield complete solution. This solution is diluted to approximately 50 ml with distilled water and is made up to 1·1 M HCl by dropwise addition of 6 M HCl, using a Beckman pH meter and a standard 1·1 M HCl solution for comparison.

This solution is passed through a 25 ml resin volume Dowex-50 ion exchange column about 10 in. long and $\frac{1}{2}$ in. in diameter. The flow rate is adjusted to about 1 ml/min. After introduction of the sample to the column, elution is continued with 1·1 M HCl. Be^{+2} is completely eluted with about 6 or 7 resin volumes (about 175 ml) of 1·1 M HCl. Al^{+3} is held on the column, but

would start to elute at about from 12 to 15 resin volumes of 1·1 M HCl. After the Be^{+2} is completely eluted, the Al^{+3} is eluted with about 100 ml of 3 M HCl and is saved for step 23.

(j) *Specific beryllium purification*

(20) The solution containing Be^{+2} is evaporated to dryness on a steam bath. The $BeCl_2 . 4H_2O$ is dissolved in 25 ml of 2 M HCl. This solution is passed through a 5 ml Dowex-1 ion exchange column 2 in. long and $\frac{1}{2}$ in. in diameter. After introduction of the sample solution the column is washed with 10 ml of 2 M HCl. The washing is combined with the first eluate. Pb^{+2} is adsorbed on the column. The Pb^{+2} may be eluted with 50 ml of 10 M HCl and saved for future work, if desired.

(21) The eluate containing Be^{+2} is again evaporated to dryness. It is dissolved in 25 ml of 0·1 M $H_2C_2O_4$–0·15 M HCl (pH = 0·9, as determined with the Beckman pH meter). This solution is passed through a 10 ml Dowex-1 column 4 in. in length and $\frac{1}{2}$ in. in diameter at a flow rate of 1 ml/min. Elution is continued with 0·1 M $H_2C_2O_4$–0·15 M HCl. Residual Al^{+3} present forms an oxalate complex and is adsorbed on the column. Be^{+2} is only slightly adsorbed and is eluted completely with 5 resin volumes (about 50 ml) of the eluting solution after the initial sample introduction.

(22) The eluate containing Be^{+2} is treated with NH_4OH at pH ~7 to precipitate $Be(OH)_2$ and is filtered on Millipore HA filter paper in a 6·5 cm Büchner funnel.

The precipitate is ignited to BeO in a platinum dish at 1000°C. The BeO is mounted on an aluminium counting dish in the usual manner, using a dust mask to prevent inhalation of the very toxic BeO dust.

(k) *Specific aluminium purification*

(23) The Al^{+3} solution from step 19 is reduced in volume to 5 ml on the hot plate. The solution is cooled in an ice bath to a temperature not greater than 10°C. A volume of 5 ml of 12 M HCl is added to the solution to bring it above the constant-boiling-mixture concentration. Dry HCl gas from a cylinder is slowly bubbled through the solution for 5 min. Ethyl ether (10 ml) is added and bubbling with HCl gas is continued with the beaker in the ice bath until the two layers disappear; this indicates that the solution is saturated with HCl. This solution is allowed to stand in the ice bath for 1 hr while $AlCl_3.6H_2O$ precipitates. The fine-grained precipitate of $AlCl_3.6H_2O$ is filtered, using suction, on a funnel having a 6·5 cm fritted glass disk. A 1:1 12 M HCl–ethyl ether solution saturated with HCl gas and cooled to 10°C is used to wash the precipitate.

The $AlCl_3.6H_2O$ is dissolved in 5 ml of distilled water and reprecipitated twice more as above. The final precipitate is dissolved in distilled water.

(24) $Al(OH)_3$ is precipitated with NH_4OH at the methyl red end-point. The $Al(OH)_3$ is filtered on a Millipore HA filter paper in a 6·5 cm Büchner funnel.

The precipitate is ignited to Al_2O_3 at 1000°C in a platinum dish. The sample is mounted in an aluminium dish in the usual manner for counting.

D. Procedure for Silicate Materials

(a) *Sample preparation and solution*

(1) The specimen is rinsed with acetone to remove any lacquer which may have been used to preserve it.

(2) A 50–150 g sample of the specimen is ground to a fine powder on an electrolytic iron sheet using an iron roller. This fine powder is transferred to a 1 l. polyethylene beaker which is placed in a water bath containing water at room temperature.

(3) The sample is dissolved in the hood by the cautious addition of 48% hydrofluoric acid. The slurry is stirred with a polyethylene stirring rod. A total of about 5 ml of hydrofluoric acid per gram of sample is used. Violent effervesence is prevented by the use of the water bath for cooling and the slow addition of the hydrofluoric acid. The mixture is allowed to stand at room temperature for 3–4 hr with occasional stirring.

The beaker containing the sample mixture is then heated in the water bath at 100°C with

occasional stirring until the mixture goes just to dryness. If large amounts of sample are being dissolved, the evaporation is speeded up by passing a stream of warm dry nitrogen over the surface of the liquid in the beaker. An additional 50 ml of hydrofluoric acid are added to the residue and it is again evaporated to dryness. A volume of 50 ml of concentrated HNO_3 is added to oxidize all Fe^{+2} to Fe^{+3} and the solution is again reduced to dryness.

(4) The residue is dissolved in 100 ml of 12 M HCl and again evaporated just to dryness to remove any remaining HNO_3 and most of the HF. This evaporation is repeated with 100 ml of 12 M HCl.

This residue from the last evaporation is dissolved in 1 l. of 9–10 M HCl and is filtered through a funnel having a 6·5 cm fritted glass disk to remove the trace amounts of insoluble residue usually encountered.

(5) An accurately known amount of Be^{+2} carrier (13·9 mg was usually used in this work) is added to the solution. Other carriers are usually not necessary because of the appreciable concentrations of aluminium and calcium in silicate materials and of iron, cobalt and nickel in stone meteorites.

(b) *Iron extraction*

(6–7) Iron is removed by solvent extraction as in Part C, steps 6 and 7.

(c) *General ion exchange separation*

(8) The aqueous solution from the iron extraction steps is then put through the general ion exchange step as in Part C, step 8. If 12 M HCl is used, Ti^{+4} is adsorbed also. It may be eluted with 9 M HCl. If the amount of iron in the sample is small, the solvent extraction step may be omitted. In this case Fe^{+3} will be adsorbed on the column in the general ion exchange step and may be later eluted with 1 M HCl. The Fe^{+3} may then be further purified by solvent extraction, if desired.

(d) *Cobalt separation and specific purification*

(9–12) Cobalt is separated and purified as in Part C, steps 9 to 12.

(e) *Separation of aluminium and beryllium from nickel, calcium and magnesium*

(13) The solution containing Be^{+2}, Al^{+3}, Ni^{+2}, Mg^{+2} and Ca^{+2} from the general ion exchange step is reduced in volume to 500 ml on the hot plate. The solution is brought to the methyl red end-point with NH_4OH. $Al(OH)_3$ and $Be(OH)_2$ precipitate at this pH, while Mg^{+2} (which is usually abundant in silicate materials) Ni^{+2} and Ca^{+2} remain in solution. The precipitates are filtered on Whatman 31 filter paper in a 12 cm Büchner funnel and washed with a 5% NH_4Cl solution.

(14) The $Al(OH)_3$ and $Be(OH)_2$ are redissolved in dilute HCl and reprecipitated three times as above with NH_4OH to assure removal of Mg^{+2}, Ni^{+2} and Ca^{+2}. The $Be(OH)_2$ and $Al(OH)_3$ are saved for step 18. The filtrates are combined and saved for the nickel and calcium separation and purifications.

(f) *Separation of nickel from calcium*

(15) Nickel and calcium are separated as in Part C, step 16.

(g) *Specific purification of calcium*

(16) Calcium is purified as in Part C, step 17.

(h) *Specific purification of nickel*

(17) Nickel is purified as in Part C, step 18.

(i) *Separation of aluminium from beryllium*

(18) The $Al(OH)_3$ and $Be(OH)_2$ from Step 14 above are treated with 100 ml of concentrated NaOH and 10–20 mg of Fe^{+3} carrier and heated to boiling. After cooling, the solution containing

the aluminate and berylliate is filtered through a funnel having a 6·5 cm fritted glass disk. The precipitate containing trace impurities carried down on the $Fe(OH)_3$, is discarded or saved for future reference, if desired.

The filtrate is diluted to 250 ml and is treated with 6 M HCl to reprecipitate $Al(OH)_3$ and $Be(OH)_2$ at the methyl red end-point.

(19) These precipitates are further processed as in Part C, step 19 to separate Al^{+3} and Be^{+2}. Approximately 100 ml wet volume of Dowex-50 resin are used for each gram of Al in the sample. If the amount of aluminium in the sample is high, it is often convenient to use two or more 250 ml, 1 in. diameter ion exchange columns simultaneously.

(j) *Specific beryllium purification*

(20–22) The purification of beryllium is completed as in Part C, Steps 20–22.

(k) *Specific aluminium purification*

(23) The Al^{+3} from the Al–Be separation step is reduced in volume to 50–100 ml on the hot plate. The solution is cooled in an ice bath to about 10°C. A volume of 10 ml of 12 M HCl are added and the solution is saturated with dry HCl gas from a cylinder.

(24) A volume of ethyl ether equal to that of the solution is added and the solution is again saturated with HCl gas in the ice bath, as shown by the disappearance of the two layers. The solution is allowed to stand for 1 hr in the ice bath to allow full precipitation of $AlCl_3 \cdot 6H_2O$.

(25) The $AlCl_3.6H_2O$ is filtered on a funnel having a 6·5 cm fritted glass disk and is washed with a 1 : 1 12 M HCl–ethyl ether solution which has been saturated with dry HCl gas and cooled to 10°C. The $AlCl_3.6H_2O$ is dissolved in 25–50 ml of distilled water and reprecipitated twice as above.

(26) The final precipitate is dissolved in 100 ml of distilled water and $Al(OH)_3$ is precipitated by addition of NH_4OH to the methyl red end-point. The $Al(OH)_3$ is filtered on Whatman-31 filter paper in a 6·5 cm Büchner funnel and ignited at 1000°C in a platinum dish. The Al_2O_3 is mounted in an aluminium dish for counting in the usual manner.

E. Recycle Procedures

(a) *Cobalt*

(1) The CoS is dissolved in aqua regia and evaporated to near dryness three times with 25-ml portions of 12 M HCl to remove the excess HNO_3. Care is taken not to evaporate the solution to dryness, since $CoCl_2$ will sublime.

(2–3) A volume of 25–50 ml of 12 M HCl is added to the small volume of solution from the last evaporation, and this solution is passed into a Dowex-1, anion exchange column as in Part C, steps 8 and 9.

(4–6) The cobalt is then solvent extracted as in Part C, steps 10 to 12.

(b) *Beryllium*

(7) The BeO is transferred to a beaker and treated with a mixture of 15 ml concentrated HNO_3 and 15 ml 9 M H_2SO_4. The mixture is boiled on the hot plate for 1 hr, or until solution is complete. The solution is diluted to 100 ml with distilled water.

(8) About 10 ml of Fe^{+3} carrier is added to the solution and $Fe(OH)_3$ and $Be(OH)_2$ are precipitated with NH_4OH at pH ~7. These hydroxides are filtered on Whatman-31 filter paper in a 6·5 cm Büchner funnel.

(9) The filter paper and the hydroxides are transferred to a 250 ml beaker and treated with 15 ml of 8 M NaOH solution. This solution is heated to boiling and filtered while warm through a funnel having a 6·5 cm fritted glass disk. The $Fe(OH)_3$ remaining on the filter is washed with 10 ml of 8 M NaOH. The washings are added to the above filtrate. The $Fe(OH)_3$ precipitate is discarded.

(10) The combined filtrate containing the berylliate in solution is diluted to 100 ml and treated with 6 M HCl to precipitate $Be(OH)_2$ at pH ~7. This precipitate is filtered on Millipore HA filter paper in a 6·5 cm Büchner funnel, dissolved in 25 ml of 6 M HCl, and reprecipitated twice with NH_4OH at pH ~7, to assure removal of the Na^+ present.

(11) The final $Be(OH)_2$ precipitate is dissolved in 25 ml of 12 M HCl and passed through a 10 ml Dowex-1 anion exchange column 4 in. long and $\frac{1}{2}$ in. in diameter. The column is rinsed with 20 ml of 12 M HCl after introduction of the sample.

(12) The eluates containing Be^{+2} are reduced in volume to 10 ml on the hot plate. Distilled water (20 ml) is added to the solution to make it approximately 2 M HCl. This solution is passed through a 10 ml Dowex-1 anion exchange column and the column is rinsed with 20 ml of 2 M HCl.

(13–14) The eluates containing Be^{+2} are evaporated to dryness. The $BeCl_2.4H_2O$ is dissolved in 25 ml of 0·1 M $H_2C_2O_4$–0·15 M HCl and further processed as in Part C, steps 21 and 22.

(c) *Aluminium*

(15) The Al_2O_3 is fused in a platinum dish with ~2 g of sodium tetraborate per gram of Al_2O_3 at 1000°C for from 4 to 5 hr. The fusion cake is dissolved in dilute HCl.

(16) The $Al(OH)_3$ is precipitated with NH_4OH at the methyl red end-point. The precipitate is filtered on Whatman-31 filter paper (or Millipore filter paper, if the amount of $Al(OH)_3$ is small) in a 6·5 cm Büchner funnel. The $Al(OH)_3$ is dissolved in dilute HCl and reprecipitated twice more as above to remove the sodium tetraborate.

(17–18) The final $Al(OH)_3$ precipitate is dissolved in 12 M HCl and reduced in volume to a volume corresponding to from 5 to 10 ml for each gram of aluminium present (10 ml minimum volume) and processed as in Part C, steps 23 and 24.

F. Discussion

After each isolation or recycle the chemical yield is determined by weighing the sample as prepared for counting and comparing with the known content of the element in the material processed (for example, Tables 2 and 3) or the weight of carrier added. Chemical yields of carriers in blank runs were: Be, 96 per cent; Al, 65–95 per cent; Co, 95 per cent. Actual yields were not always as good as the blank-run values.

The apparent beryllium yields from meteorites and tektites were sometimes over 100 per cent, because of incomplete purification from bulk constituents or because of amounts of beryllium present in the meteorites which were not negligible in comparison to the amounts of carriers added. That the former cause is mainly responsible appears probable from the large decrease in weight of "BeO" samples often observed on recycling, and from the high initial yields of "BeO" from iron meteorites. If the beryllium content of the stone meteorite specimens is close to the accepted average for these objects, ~1·2 p.p.m. (Suess and Urey, 1956), samples weighing several hundred grams should contain less than 1 mg of the element; however, individual objects may have more than the average, and tektites might conceivably have unexpectedly high beryllium contents, in which case the second cause of high yields may also be important.

Many samples of BeO, Al_2O_3, and CoS have been prepared from meteorites and terrestrial materials and measured with the specially constructed low-level beta counters described in the following sections of this paper. Early procedures used during the course of development yielded some samples of moderately high activity, which often exhibited growth or decay, or which fell considerably on a specific-activity basis upon recycling. Uranium and thorium and their disintegration products are the most likely radioactive contaminants. The final procedures for aluminium and cobalt are quite satisfactory. With these elements the initially isolated sample was usually either completely inactive, or exhibited

an activity which changed very little on a specific-activity basis upon recycling and which did not change with time.

The beryllium procedure has not been carried to quite as satisfactory a state, even though it should be specific for that element. The initial activity usually decreased on recycling. It appears that at least three recycles may be necessary to get rid of all contaminating activities and inert impurities. The radioactive contamination does not grow or decay perceptibly in periods of days to months, and its chemical or nuclear identity cannot be surmised. Since reagent beryllium, like reagent aluminium and reagent cobalt, showed no activity after being put through these procedures, the contamination is almost certainly associated with the natural radioelements present in the samples. Further development of the beryllium purification part of the procedure would be desirable to reduce the number of recycles required. A recent beryllium separation method making use of beryllium acetylacetonate solvent extraction into CCl_4 may prove to be of value (Alimarin and Gibalo, 1956).

3. Instrumentation and Radiometric Procedures

A. Objectives and General Nature

Of the nuclides likely to be present (Table 1 and Fig. 1), most belong to elements which do not form volatile compounds, and can be measured most readily by counting their radiations from thin layers of solids. Most of these nuclides are beta emitters. Many of the elements of which they are isotopes are minor constituents in stone or in iron meteorites or in both, and accordingly can be prepared in rather small samples. Moreover, a few of these nuclides have active daughters, which can be separated from their parents with only small amounts of carrier. Accordingly, a counter having a high sensitivity for small solid beta-emitting samples should have a considerable range of usefulness in work of this kind, and was selected for our exploratory experiments.

At the time this work was begun, there were no experimental data on the rates of production of new atoms in meteorites by cosmic rays or in targets exposed to particles artificially accelerated to similar energies. However, Singer (1952, 1953) and Martin (1953) had estimated the production rates of isotopes of helium and hydrogen. Singer calculated that in the region 2–15 cm below the surface of a large iron meteorite the total He production rate should be $(5 \pm 1{\cdot}5) \times 10^{-14}$ $cm^3\ g^{-1}\ yr^{-1}$, corresponding to $\sim$2·5 atoms $min^{-1}\ g^{-1}$. Of the latter, $\sim$1·7 would be He^4, $\sim$0·3 He^3 produced directly, and $\sim$0·5 He^3 produced indirectly through H^3. The latter figure gives the expected saturation radioactivity of H^3, $\sim$0·5 dis $min^{-1}\ g^{-1}$. Martin's formulas yield for the outer 20 cm of a large iron meteorite (depending on depth) 3·0–3·8 atoms $min^{-1}\ g^{-1}$ as the combined production rate of He^4, He^3 (direct) and H^3. These estimates have been confirmed roughly by subsequent measurements of H^3 production rates in BeV proton bombardments using accelerators and of the H^3 activities in meteorites. For specific heavy atomic reaction products one might expect the yields to be lower by factors of the order of ten or a hundred. One could expect, then, activities of individual radionuclides of the order of 10^{-1} or 10^{-2} dis $min^{-1}\ g^{-1}$, or even lower if shielding, decay, or unsaturation were important.

Accordingly, even if meteorite sample sizes of tens or hundreds of grams were used, it would be necessary to be able to detect isolated activities of the order of a fraction of a count per minute; accordingly, conventional types of counters, having backgrounds of several counts per minute, would not be satisfactory. Besides the statistical fluctuations in counting rates, there are real fluctuations in the backgrounds of such counters, due to variations in the local cosmic-ray intensity, amounting to some 5 per cent. These effects set a lower limit of several counts per minute for the useful range of activities measurable with ordinary counters. Therefore, reduction of the background, especially of the cosmic-ray contribution by anticoincidence, is essential. The resulting reduction of the real background variations is probably a more important factor than the reduction of the random variations due to statistical fluctuations.

Recent reviews of the general subject of low-level radioactivity measurement have been given by JOHNSTON (1956) and by ANDERSON and HAYES (1956). Other counters similar to ours have been described (VOLCHOK and KULP, 1955; GRUMMITT *et al.*, 1956; SOFTKY and NATHER, 1957; LAL *et al.*, 1957) but we believe that several aspects of the construction and performance of our system merit a brief description.

B. Low-level Beta Counting System

(a) *Side-window Geiger counters*

For small samples the geometry factors of window-type counters can be made nearly as high (~0·3–0·4) as the maximum possible with internal sample counters (0·5). External-sample counters have the advantages of greater stability, easier changing of samples, and less danger of contamination. Since all of these factors are important for routine counting of weak samples over long periods, we adopted the window-type arrangement. A side-window straight-wire design was chosen because such a counter would have a higher ratio of window area to gas volume than end-window counters, it would have no dead-space as in bead-terminated straight-wire end-window counters, and it should operate more reliably in the Geiger region than loop-anode end-window counters. The size was a compromise between the demands of low background (requiring a small volume) and high geometry factor (requiring a large window).

It is necessary to construct the counter tubes themselves out of materials which have little radioactivity. A survey of the amount of radioactivity found in ordinary apparatus construction materials has been given by GRUMMITT *et al.* (1956). Of these materials, the plastic Lucite showed one of the lowest activities. For this reason, the counters were constructed of Lucite, and materials known to have high radioactivities, such as solder, glass, brass and mica, were avoided.

Fig. 2 illustrates the construction details of these counters. The active length of the tube is 4·8 cm and the inner diameter 1·27 cm tungsten wire (0·025 mm) is used for the anode, and a layer of Aquadag, a graphite suspension in water, is painted on the inner surfaces to serve as the cathode. DeKhotinsky cement is used to seal the holes for the centre wire, and steel screws are used for the electrical binding posts.

The window is 0·97 mg cm^{-2} Mylar film coated with aluminium. Its area is

slightly over 6·0 cm². The foil, somewhat larger, is affixed to the Lucite tube with rubber cement to obtain a reasonably gas-tight seal. Since the counter is operated as a flow counter at essentially atmospheric pressure, no elaborate gas-tight seals are necessary. Electrical contact between the Aquadag cathode, the aluminized inner surface of the window, and the cathode terminal is achieved by painted Aquadag connexions and a thin strip of aluminium foil.

The choice of operation as Geiger rather than proportional counters was mainly based on the simplicity of the circuitry for Geiger operation. The counters are operated as flow counters using as a counting gas a 98·7% helium–1·3% butane

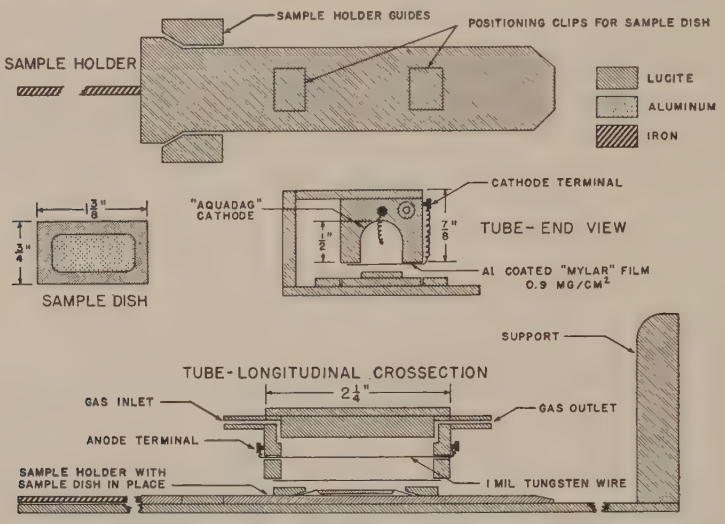

Fig. 2. Side-window small-sample low-level Geiger counter with sample holder and positioning mechanism.

gas mixture("*Q*-gas"). The gas flow rate is adjusted using a bubbler to about 100 small bubbles/min for the bank of four sample counters connected in parallel.

The counters operate at 1125 V d.c. on a plateau of approximately 150 V in length with a slope of 5 per cent per 100 V. The height of the pulse delivered at this voltage is approximately 0·18 V.

(b) *Sample mounting*

In the interests of long-term stability and freedom from contamination, it was deemed necessary to be able to insert and remove samples from the shield without disturbing the counter inside. Fig. 2 shows also the sample-holding mechanism by which this was accomplished.

Standardized sample dishes 1·9 × 3·5 cm² in outer dimensions were made out of 0·004 mm thick aluminium foil. In each was stamped a depression 1–2 mm deep, 2·54 cm long and 1·14 cm wide with rounded corners, so that the area was 2·88 cm². This can accommodate samples of size up to that of 400 mg Al_2O_3. Before use each dish is boiled in ethyl alcohol to remove grease and other dirt. The filled sample depression is then circled, using a camel's hair brush, with rubber cement diluted 1 : 1 with toluene. A strip of cellophane from a standard

supply is wiped on both sides with a piece of cotton saturated with ethyl alcohol. This strip is then applied to the top of the counting dish and sealed by pressing it into the rubber cement. The excess cellophane is trimmed off and the entire outer surface of the dish is again wiped with a piece of cotton saturated with ethyl alcohol.

The dish is placed on the Lucite sample holder and centred under the positioning clips. The holder is then pushed under the counter by removing one brick from the end of the shield. The sample holder positions itself reproducibly under

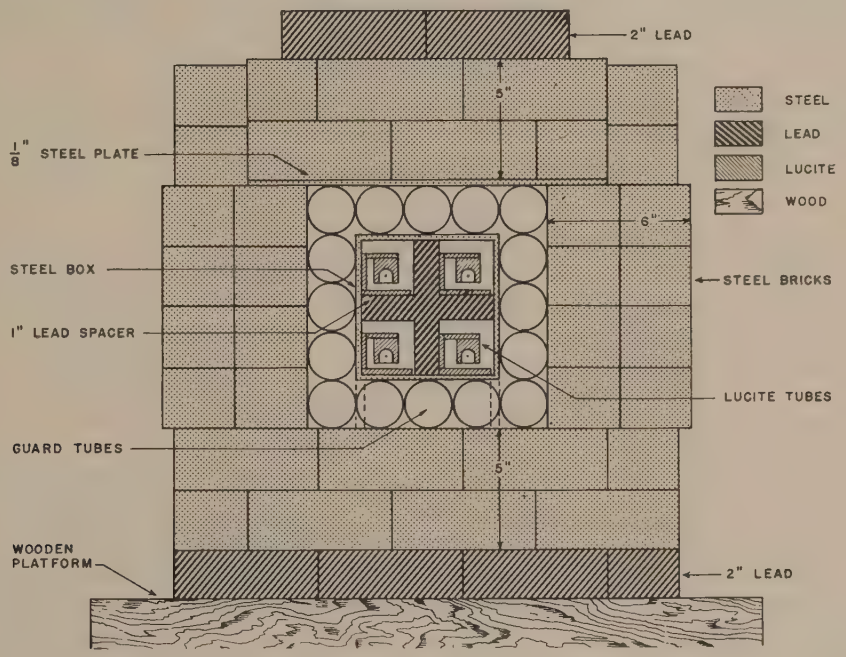

Fig. 3. Multiple low-level beta counter assembly with guard counters and shield.

the counter by virtue of the locking effect of the sample-holder guides. The counter itself need only be removed from the shield for occasional repair.

(c) *Multiple-counter assembly*

Because of the long counting times necessary for each sample and background, it was found essential to have several counters which could be operated simultaneously and essentially independently. For economy and convenience, four identical sample counters were mounted inside the same sheath of guard counters and shield. The arrangement is shown in Fig. 3.

The four counters are supported in a $15 \times 15 \times 25$ cm^3 box constructed of 3 mm thick steel. They are separated from each other by 2·5 cm thick plates of lead arranged in the form of a cross within the steel box. These spacers prevent a sample positioned below one counter from affecting the other three counters. 500 disintegrations min^{-1} of Na^{22} placed under one sample counter did not perceptibly increase the backgrounds of the other counters.

The guard tubes for anticoincidence background reduction are of a common type of Geiger counter used in cosmic ray research. These tubes are made of brass and have an over-all length of 38 cm and an outer diameter of 5·1 cm. The active length of the tubes is 33 cm and the wall thickness is 0·8 mm. They are filled with an argon–alcohol mixture at about 16 cm pressure. Sixteen of these tubes connected in parallel are arranged in a square array around the sample-counter supporting box. The normal counting rate of the anticoincidence bank in the steel shield is 1150 counts min^{-1}.

The massive shield surrounding the guard tubes is constructed of steel and lead bricks.

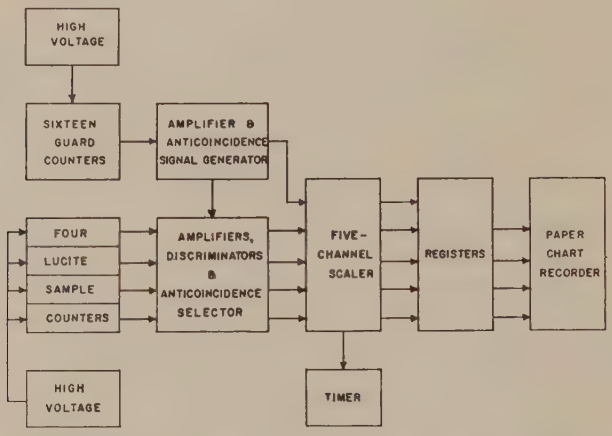

Fig. 4. Block diagram of anticoincidence counting and recording system.

A block diagram of the entire counting system is shown in Fig. 4. The details of the circuitry and the recording system are discussed elsewhere (Ehmann, 1957).

C. *Backgrounds and Counting Procedure*

The sample counters have unshielded backgrounds of about 11 counts min^{-1}, which is reduced to about 6 counts min^{-1} by the massive shield. The anticoincidence guard tubes reduce this further to between 0·35 and 0·45 countsmin^{-1}. With an argon–ethylene counter gas instead of the helium-rich gas finally adopted, the lowest backgrounds were about 0·8 counts min^{-1}; this is due to the greater gamma sensitivity of argon, and illustrates the importance of using a gas of low atomic number.

There was very little evidence for real variations in the background correlated with the atmospheric pressure. For the long counting periods used, cyclic variations of barometric pressure would cause any such effect to average out.

As a result of the elimination of most of the cosmic-ray component of the background, of the freedom from contamination resulting from the permanent mounting of the counters inside the shield, and of the inherent stability of flow-type counters, the backgrounds were remarkably stable over long periods of time (see Ehmann, 1957).

The following is the normal procedure used for counting samples and

backgrounds: (1) An empty sample dish is counted for 3 or 4 days; (2) the dish containing the sample is counted for 6 to 8 days; (3) the empty dish is again counted for 3 or 4 days. Step (3) is used as step (1) in the counting cycle for the next sample. The backgrounds immediately preceding and following each sample count are averaged and subtracted from the latter. Accordingly, both gross and background rates are based on 6–8 day counts, but each background counting period can be used twice. With all counters operating simultaneously, complete counting data for four samples are obtained every 9–12 days. The continuous monitoring of the backgrounds is a necessary but convenient check on the functioning of the counters.

No detectable change in the background was observed with the aluminium sample dishes and cellophane covering foils used. Freedom from noise and electrical pickup was demonstrated by operation with the centre wires of the sample Geiger counters disconnected; no pulses were recorded in 48 hr.

D. *Counting Yields for Thin Sources*

A counting yield, Y, may be defined as the ratio of the net counting rate (corrected for background) to the absolute disintegration rate. The counting yield may be expressed as a product of several more-or-less independent factors (for example, STEINBERG, 1956):

$$Y = \frac{\text{net counting rate}}{\text{absolute disintegration rate}} = G.W.A.C.B.H.S.I$$

where G = geometry factor
W = factor for the absorption effect of the counter window and the air between the source and the window
A = factor for the scattering effect of the air.
C = factor for the effect of any covering material over the sample
B = factor for the effect of backscattering by the material supporting the sample
H = factor for the effect of the sample and the tube supporting structure and the walls of the shield in scattering counts into the counter
S = factor to correct for finite sample thickness
I = the detection probability (weighted average for the various radiations emitted)

In this work the first six factors were grouped together and determined experimentally as a unit, since these factors are quite complex, and it is not easy to separate them experimentally. Therefore, the counting yield equation for a sample of finite thickness, θ, reduces to:

$$Y(\theta) = Y_0 \,.\, S(\theta) \,.\, I$$

where Y_0 is the product of the first six factors. Y_0 was determined by preparing thin sources from standardized solutions of several beta emitters and counting them in a manner identical to that used for normal samples. Since S for very thin samples is by definition unity:

$$Y(\theta) = Y_0 \,.\, I$$

The two radionuclides Al^{26} and Na^{22} were used to obtain values for Y_0. Both are positron and gamma emitters and both also decay in part by electron capture. These radionuclides were obtained in carrier-free form from standardized solutions prepared by Rightmire of this laboratory. The solutions had been standardized by means of a 4π beta counter operated in the Geiger region (Rightmire 1957). The absolute beta disintegration rates of these samples were known to about ± 2 per cent.

(a) *Experimental determination for* Al^{26}

An aliquot of the standardized Al^{26} solution was evaporated on a standard counting dish, covered with cellophane in the usual manner, and counted under one of the side-window counters. The ratio of the observed counting rate to the known absolute positron emission rate (Y_0 for Al^{26}) was found to be $0{\cdot}49 \pm 0{\cdot}01$. The detection probability factor, I, for Al^{26} is 0·84, based on a fractional electron capture of 16 ± 2 per cent (Rightmire *et al.*, 1958). These counters are insensitive to electron-capture radiations and nearly insensitive to gammas, while the detection probability for betas is assumed to be very close to unity. Therefore, the counting yield equation for Al^{26} is:

$$Y(Al^{26}) = Y_0(Al^{26}\beta^+) \,.\, I(Al^{26}) \,.\, S(\theta) = (0{\cdot}49)(0{\cdot}84)S(\theta) = 0{\cdot}41\, S(\theta)$$

(b) *Experimental determination for* Na^{22}

Since standardized samples of Be^{10} and Co^{60} were not available, Na^{22} was used to obtain an indication of the energy dependence of Y_0 for betas. Although Be^{10} and Co^{60} are negatron emitters, whereas Na^{22} and Al^{26} are positron emitters, it was assumed that the difference in the backscattering factor, B, for negatrons and positrons of the same energy is not great enough to be of significance here.

An aliquot of the standardized Na^{22} solution was counted under the same counter as was used for the Al^{26}. The counting yield for Na^{22} positrons was found to be $0{\cdot}44 \pm 0{\cdot}01$. The lower value (0·44) for Na^{22} ($E_{max} = 0{\cdot}54$ MeV) than that (0·49) for Al^{26} (1·16 MeV) is presumably due to a smaller backscattering factor for the lower energy positrons. Since the electron-capture branching fraction of Na^{22} is 11 per cent

$$Y(Na^{22}) = Y_0(Na^{22}\beta^+) \,.\, I(Na^{22}) \,.\, S(\theta) = (0{\cdot}44)(0{\cdot}89)S(\theta) = 0{\cdot}39\, S(\theta)$$

(c) *Estimation for* Be^{10} *and* Co^{60}

The maximum energy of the Be^{10} negatrons (0·56 MeV) is very close to that of the Na^{22} positrons (0·54 MeV). Accordingly, neglecting small differences in backscattering and in spectrum shape effects, we can assume that $Y_0(Be^{10}\beta^-) = Y_0(Na^{22}\beta^+)$. Therefore:

$$Y(Be^{10}) = Y_0(Na^{22}\beta^+) \,.\, I(Be^{10}) \,.\, S(\theta) = (0{\cdot}44)(1{\cdot}00)\, S(\theta) = 0{\cdot}44\, S(\theta).$$

Co^{60} is a negatron emitter of somewhat lower maximum energy, 0·31 MeV. Considering the energy dependence as determined by Al^{26} and Na^{22} and neglecting minor effects, we adopt:

$$Y(Co^{60}) = (0{\cdot}42)(1{\cdot}00)\, S(\theta) = 0{\cdot}42\, S(\theta)$$

E. *Sample-thickness Factors*

The sample thickness factor, $S(\theta)$, differs from unity for finite thicknesses because of the combined effects of self-absorption and self-scattering. The function is different for each nuclide and for each set of measuring conditions, depending strongly on the maximum energy of the beta particles. So far, only experimental evaluation has proven satisfactory; for this there are two principle techniques (KOHMAN, 1949): (1) measurement of a constant amount of activity in various thicknesses of sample, and (2) measurement of various thicknesses of sample of constant specific activity.

(a) *Experimental determination for* Al^{26} *in* Al_2O_3

Since Al^{26} of high specific activity was available through the courtesy of RIGHTMIRE (see RIGHTMIRE *et al.*, 1957), method (1) could be used. This method has the advantage of giving $S(\theta)$ directly, without the necessity of an extrapolation to $\theta = 0$.

Aliquots of the same Al^{26} solution used in the determination of the thin-sample counting yield (Section 3Da), were added to various accurately known weights of carrier aluminium in solution. These carrier solutions were prepared by dissolving a weighed piece of 99·99 per cent pure Al metal in hydrochloric acid. This solution was diluted to 50 ml with distilled water. Different volumes of this solution were accurately pipetted into small beakers to which the Al^{26} aliquots were added. The aluminium samples were then precipitated with NH_4OH at the methyl red end-point and ignited to Al_2O_3 in individual porcelain crucibles. The chemical yield was determined for each sample by weighing the Al_2O_3. These samples were then mounted for counting in the usual manner. An equal aliquot of carrier-free Al^{26} was evaporated on a standard aluminium dish and covered with cellophane in the usual manner.

The counting rates of these samples were determined with the window-type counter. The ratio, $S(\theta)$, of the experimental counting rates of each sample to the counting rate of the carrier-free sample (corrected for the chemical yield of each sample) was then plotted against sample thickness, θ (mg cm^{-2}). This plot is given by Fig. 5. The shape of this curve conforms roughly to those obtained for typical beta emitters by NERVIK and STEVENSON (1952). However, the maximum is less pronounced than for the curve for 1·16 MeV obtained by interpolating among those authors' curves for NaCl samples, mainly because of the lower effective atomic number and scattering power of Al_2O_3.

(b) *Experimental determination for* Be^{10} *in* BeO

For this there was available only a source of Be^{10} of low specific activity, so method (2) had to be used.

A Be^{10} solution was prepared from a beryllium target which received a long bombardment with deuterons at the University of Pittsburgh cyclotron, and which was kindly supplied by Professor A. J. ALLEN. The reaction producing Be^{10} is Be^{9}(d,p)Be^{10}. The target had decayed for several years after removal from the cyclotron. Therefore, any 54-day Be^{7} produced would have disappeared. The

sample was purified by repeated precipitations of $Be(OH)_2$ with NH_4OH at pH 7 as determined with Alkacid test paper. Sodium carrier was used in these precipitations to carry out any Na^{22} present. An absorption curve on the purified BeO showed a pure 0·56 MeV beta.

Three BeO samples of different weights were prepared from the above solution. These samples were weighed and mounted in the usual manner and counted under the window-type counter. A plot of the experimentally observed specific counting rate versus the sample-thickness was prepared. This plot was extrapolated back

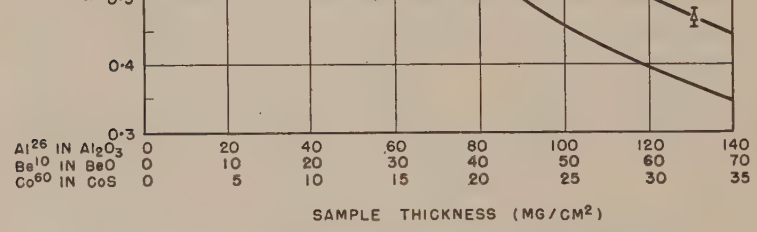

Fig. 5. Sample-thickness factors for Al^{26}, Be^{10} and Co^{60}.

to zero sample-thickness, conforming to the shape of curves given by Nervik and Stevenson for beta emitters of similar energies (but with a much flatter maximum corresponding to the much lower effective atomic number of BeO), to obtain a value of the carrier-free specific counting rate of the Be^{10}. The ratio, $S(\theta)$, of the finite-thickness specific counting rates to the carrier-free specific counting rate as determined above was plotted against θ. This plot is also shown in Fig. 5.

(c) *Estimation for* Co^{60} *in* CoS

Sample thickness factors for Co^{60} were not determined. A plot of $S(\theta)$ versus θ for a typical 0·3 MeV beta emitter in NaCl as given by Nervik and Stevenson and extrapolated to greater sample thicknesses is also shown in Fig. 5, and was used for evaluation of our Co^{60} measurements. Here we have neglected the effect of the appreciably higher effective atomic number of CoS as compared to NaCl.

F. *Sensitivity of Counters*

Since the backgrounds are stable over long periods and fluctuate only in the manner and degree expected for random events, the sensitivity is determined only by the statistical fluctuations and the counting yields. We regard here, as in other work in this laboratory (Selig, 1954; Diethorn, 1956), that the lower limit of the counting rate that may be reliably detected is twice the standard deviation σ, of the difference between the gross sample count and the background count. There is a probability of 0·977 or close to certainty, that any net counting rate exceeding twice its standard deviation represents a real radioactivity in the sample.

Since the backgrounds of these counters were close to 0·4 count min^{-1}, for gross counts only slightly in excess of background $2\sigma = 0{\cdot}020$ count min^{-1} for the difference between 6-day counts of both sample and background, and $2\sigma = 0{\cdot}017$ count min^{-1} for 8-day counts. Since the counting yields for the nuclides measured are usually close to 0·4 count dis^{-1}, lower limits to the absolute activities detectable are about 0·04 dis min^{-1}.

Since specimen sample sizes of about 100 g are usually used, lower limits to the specific activities are therefore approximately 0·0004 dis min^{-1} (g specimen)$^{-1}$ in favourable cases where most of the radionuclide can be concentrated into a small sample for counting. Such cases are Be^{10} in all meteorites, Al^{26} in irons, and Co^{60} in stones. Higher limits result for such cases as Al^{26} in stone meteorites and most terrestrial silicates and Co^{60} in iron meteorites, since only a part of the Al_2O_3 or CoS obtained from such large samples can be counted with the present arrangement.

G. *Discussion*

This counting system has proved to be an extremely reliable and useful piece of equipment in our laboratory. It has been effective in the measurement of Al^{26}, Be^{10} and Co^{60} isolated from some meteorites and tektites (Ehmann and Kohman, 1958). In this work counting rates of samples believed to be reliably purified ranged from $\sim$0·2 count min^{-1} down to the sensitivity limit, $\sim$0·02 count min^{-1}.

An important feature of the system is the mechanism for inserting and removing samples through a small opening in the shield without touching the counters or disturbing the flow of gas or the applied voltage. This probably contributes significantly to the long-term stability of the backgrounds.

The backgrounds could probably be lowered further by replacing the lead spacers between the counters by mercury and surrounding the counter box inside the guard counters with a mercury shield. Neutron shielding (Devries, 1956, 1957) would also help. With such modifications backgrounds of 0·1 count min^{-1} should be obtainable; this would double the sensitivity.

Besides Al^{26}, Be^{10} and Co^{60}, these counters should be useful for measurement of Cl^{36}, C^{14}, Si^{32}, Na^{22}, and several shorter-lived radionuclides from meteorites. In addition, Fe^{60}, Ti^{44} and Si^{32} could be effectively measured through extraction of their active daughters Co^{60}, Sc^{44}, and P^{32}, respectively, thus overcoming the difficulties associated with the low specific activities of Fe^{60} in all meteorites and of Ti^{44} and Si ^{32} in stone meteorites.

A serious limitation for many nuclides of interest is the requirement of small sample size. Although larger counters of the same type would be useful, it would be better to use cylindrical thin-wall tubes which could be completely surrounded by the sample layer. Small foil-wall flow counters of the type developed by Sugihara *et al.* (1953) have been successfully used in low-level applications (Libby and Schrodt, 1954; Ely *et al.*, 1955). These should have high sensitivity for samples up to several grams. We constructed and tested such a counter having a diameter of 1·27 cm and an active length of 4·8 mm, but found that it exhibited unexpected and unexplained background variations. One of their drawbacks is that they must be removed from the shielding pile in order to change samples, whereas the window-type counters described above may be left in the shield.

In addition to the usefulness of this counting apparatus in detecting and measuring low levels of radioactivity, its recording system would make it very useful in following the decay of short-lived activities without the necessity of constant attention. This would be valuable for the study of short-lived activities in freshly-fallen objects as well as of P^{32} and Sc^{44} isolated from their long-lived parents.

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Cosmic-ray-induced radioactivities in meteorites—II
Al^{26}, Be^{10} and Co^{60}, aerolites, siderites and tektites*

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Abstract—The long-lived radionuclides Al^{26} and Be^{10} and the 5 year Co^{60} have been detected in stone and iron meteorites. The highest levels observed, in disintegrations per minute per gram of specimen were: in stone, Al^{26}, 0·063; Be^{10}, 0·0051; Co^{60}, 0·0018; in iron, Al^{26}, 0·0055; Be^{10}, 0·00070; Co^{60}, 0·033. The Al^{26} and Be^{10} were evidently produced by cosmic-ray bombardment of the meteorites before fall. The Co^{60} seems to have been produced principally by neutron capture in ordinary cobalt in recent years while the specimens were on earth, although disintegration of the long-lived Fe^{60} may contribute some Co^{60}.

The Be^{10} : Al^{26} ratio is definitely lower in the Plainview chondrite than in the Richardton chondrite, suggesting that the former may have existed as a small body for only about a million years. The presence of both activities in the Odessa siderite indicates that it and its associated craters are probably less than about a million years old. The absence of these activities in the Canyon Diablo siderite is probably ascribable to a shielded position of the specimen inside the large meteoroid.

Al^{26} is definitely and Be^{10} fairly certainly present in australites, indicating that tektites are glass meteorites and have spent at least a million years or so in space. Both activities are also present in Libyan Desert silica-glass, indicating that it is a variety of tektite. From the Al^{26} and Be^{10} levels or limits, in comparison with the levels in chondrites, the following approximate terrestrial ages or limits, in millions of years, are calculated: australites, $<0·5$; Libyan Desert silica-glass, $\sim 3·5$; bediasites, $>1·2$; moldavites, >3.

Measurements of these and other nuclides can give valuable information on the pre-fall and post-fall histories of meteorites and on the past intensity of the cosmic radiation. The average cosmic-ray flux over the past several million years appears not to have been grossly different from the present value.

1. Introduction

The previous paper in this series (Ehmann and Kohman, 1958) has been concerned with the chemical separation techniques and the instrumentation developed for the detection of certain cosmic-ray-induced radioactivities in meteorites. The potential usefulness of these studies and the previous work in this field have been discussed elsewhere (Kohman and Ehmann, 1958).

For reasons discussed in Paper I, the long-lived nuclides Be^{10} ($H = 2·7 \times 10^6$ years) and Al^{26} (8×10^5 years) and the 5 year Co^{60} (the daughter of the long-lived Fe^{60}, $\sim 3 \times 10^5$ years) were selected for this exploratory investigation, and this paper is concerned only with these three nuclides. The preliminary results (Ehmann, 1957), which have been presented briefly elsewhere (Kohman and Ehmann, 1958; see also Friedman, Kohman and Cassidy, 1958), are modified slightly and supplemented by some additional data in this paper.

2. Materials

We have sought for investigation representative specimens of stone meteorites, iron meteorites, tektites, and the unique Libyan Desert silica-glass. It was hoped that the presence or absence in tektites and Libyan glass of induced activity at levels comparable to those in acknowledged meteorites would throw light on the view held by many that the tektites and Libyan glass are extra-terrestrial in origin, constituting a class of "glass meteorites". For this

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purpose, control measurements on genuinely terrestrial materials are necessary, to prove the absence in them of the same activities at comparable levels. Some activation should result from the small fraction of the cosmic-ray primaries which penetrate the earth's atmosphere and from the secondaries at the surface, particularly the "nucleonic component". In special cases, slow-neutron-capture activation may also be important. Some nuclear reactions can also be induced by the alpha particles and neutrons associated with uranium and thorium in or near the specimen. In all cases, the rates of terrestrial production by cosmic rays should show a moderate dependence on geomagnetic latitude and a strong dependence on altitude above sea-level. Accordingly, in the following descriptions we record, in addition to the geographical coordinates, the altitude above sea-level of the place of collection of each specimen, as well as can be determined.

A. Terrestrial Materials

Ordinary analytical grade reagent chemicals of Al, Be and Co were used for testing the procedures and for carriers when the latter were required.

To test the possibility of activation of Co^{60} by neutron capture by cobalt at the earth's surface (Davis and Schaeffer, 1955) several terrestrial samples of cobalt were examined: *Tech cobalt*—this refers to the above mentioned reagent, a bottle of $CoCl_2.6H_2O$ which evidently had been in a basement storeroom at this Institute (lat. 40°27′ N, long. 79°57′ W, elev. 280 m) for many years. *Colorado cobalt*—this was a sample of $CoCl_2.6H_2O$ which had been in the Department of Chemistry, University of Colorado, Boulder, Colorado (40°2′ N, 105°15′ W, ~1750 m) supposedly for many years. This was kindly given to us by Prof. R. N. Keller. *Ontario cobaltite*—this was a sample of cobaltite ore from the surface of a mine dump near Cobalt, Ontario, Canada (47°14′ N, 79°25′ W, ~320 m). According to its donor, Mr. Robert Thomson, Resident Geologist, Ontario Department of Mines, the sample had probably been on or near the surface for 20 or 25 years.

The *Ontario granite* used was a piece of a porphyry boulder from the surface of the Temiskaming Conglomerate near Kirkland Lake, Ontario, Canada (48°6′ N, 79°50′ W, ~320 m). It was supplied by Prof. R. C. Emmons, Department of Geology, University of Wisconsin, Madison, Wisconsin.

The *California obsidian* was from an exposed face on Glass Mountain near Coso Hot Springs, Inyo County, California (36°2·5′ N, 117°48·5′ W, ~1290 m). It was donated by Dr. A. J. Cohen, Mellon Institute, Pittsburgh, Pennsylvania.

B. Meteorites

All of the meteorite specimens used were purchased from Ward's Natural Science Establishment, Rochester, New York. Details of their occurrence and characteristics are give by Prior and Hey (1953).

The *Richardton aerolite* fell in Stark County, North Dakota (46°53′ N, 102°19′ W, ~800 m) on 30 June 1918. It is described as a veined spherical bronzite–chondrite. About 100 kg had been collected up to 1952. Our sample was a portion of a complete individual weighing 310 g, which was entirely covered with black fusion crust.

The *Plainview aerolite* was found in many fragments in Hale County, Texas (34°7′ N, 101°47′ W, ~1100 m) shortly before 1917. It may have fallen in 1902 or 1903 (Nininger, 1952, p. 195). It is classified as a polymict brecciated veined intermediate chondrite. About 700 kg had been collected up to 1952. One sample was a complete individual which weighed 200 g; the other was part of an individual weighing 700 g. Each showed a fusion crust.

The *Odessa siderite* was found in Ector County, Texas (31°43′ N, 102°24′ W, ~900 m) before 1922. It is associated with a well-developed crater which is smaller and considerably more weathered than the Canyon Diablo crater, and with several additional much smaller craters. A coarse octahedrite, in composition and structure the metal is so similar to Canyon Diablo as to suggest to some that the two meteorites were parts of the same fall. A total of several hundred kilograms had been collected up to 1952. Our specimen was cut from a nearly complete individual weighing about 2 kg. The specimen appeared to have portions of a fusion crust.

The *Canyon Diablo siderite* is associated with the large impact crater ("Meteor Crater", "Barringer Crater") (Nininger, 1956) in Coconino County, Arizona (35°3′ N, 111°2′ W, ~1800 m). It is believed that the crater was formed about 50,000 years ago, that most if not all of the many large and small metallic fragments and oxidized residues were parts of a single massive body which entered the earth's atmosphere, and that the 30-odd tons which have been collected up to the present represent only a minute fraction of the original mass. Most specimens are coarse octahedrites. Our samples were cut from a larger individual and each had portions of an oxide crust (not a fusion crust).

The *Canyon Diablo spheroids* are small spheroidal particles, metallic except for a thin oxide coating, ranging in size from ~0·1 to ~2 mm in diameter, which occur on the desert surface surrounding the Meteor Crater to distances of several kilometres (Nininger, 1952, 1956). Their cobalt and nickel contents (see Table 3 of Paper I) are considerably greater than those of the bulk of Canyon Diablo metal, suggesting an origin by condensation of vaporized metal. The sample was purchased from Dr. H. H. Nininger, American Meteorite Museum, Sedona, Arizona, who had collected the spheroids from the top few centimetres of the ground, where they had evidently been lying for years. They were collected since 1955 at an elevation of ~1700 m and stored in a cinder-block building at ~1340 m near the site of collection.

C. Tektites and Silica-glass

The *australite* sample used was composed of about thirteen fragments from the Cook collection made in the Kalgoorlie region of Western Australia (~31° S, ~122° E, ~400 m). They were given to us by Dr H. M. Hale, Director of the South Australian Museum, Adelaide, South Australia. Detailed descriptions of australites are given by Fenner (1934, 1935, 1938) and Baker (1944). Many of the fragments used had portions of the characteristic flanges associated with australites.

The *bediasite* used was a single specimen, originally from the collection of Mr. A. C. Buscha, Giddings, Texas, and supplied by Prof. J. A. S. Adams, Department of Geology, The Rice Institute, Houston, Texas. Bediasites have been described by Barnes (1940). They are found as they weather out of the Upper Eocene Jackson sedimentary formation, mostly in Grimes County, Texas (30°36′ N, 96°6′ W, ~100 m). Our specimen had no distinctive shape and appeared to be very weathered.

Several *moldavite* individuals, each weighing from 15 to 20 g, made up the specimen used. They were obtained for us by Dr. E. F. Kohman from the widow of Herr Anton Berger of Mödling bei Wien, Austria. The most extensive description of moldavites is that of Suess (1900); a survey of more recent publications has been given by Barnes (1940). They occur mainly in Bohemia and Moravia, Czeckoslovakia (~49° N, ~14–16° W, ~500 m), in Middle Miocene Helvetien gravels. Our specimens had deeply eroded surfaces and were green to transmitted light, and therefore were presumably of the Bohemian variety.

The *Libyan Desert silica-glass* occurs in south-western Egypt between latitudes 25°2′ and 26°13′ N and longitudes 25°24′ and 25°55′ E, the elevation being ~400 m. Our specimens were collected by L. J. Spencer in 1934 and were given to us by Dr. G. F. Claringbull of the British Museum, London. Information on the discovery, occurrence, and composition is given by Clayton and Spencer (1934), Spencer (1939), Clayton (1951) and Oakley (1952). According to the latter, the glass has been in the region for at least 10,000 years and apparently many times longer. Our specimens had no characteristic shape, were a light straw-colour to transmitted light, and appeared quite sand-worn.

3. Results

The experimental data pertaining to the radioactivity measurements on aluminium beryllium, and cobalt samples isolated from the various materials are presented in Tables 1, 2 and 3, respectively, and the results are summarized in Table 4. In the first three tables the next-to-last column gives the calculated specific activity of the Al_2O_3, BeO or CoS counted. The last column gives the

specific activity of the measured radioactivity in the original specimen, as does also Table 4.

The sample specific activity is obtained by dividing the net (background-corrected) counting rate by the zero-thickness counting yield, the sample-thickness factor, and the sample weight. Zero-thickness counting yields as determined or estimated in Paper I are: Al^{26}, 0·41; Be^{10}, 0·44; Co^{60}, 0·42. Sample-thickness factors are taken from Fig. 5 of the same paper, using a sample area of 2·88 cm², and are listed in the columns headed "$S(\theta)$".

The specimen specific activity is obtained by dividing the net counting rate by the zero-thickness counting yield, the sample-thickness factor, the specimen weight, and the fraction of the element in question in the sample counted. The latter quantity, as calculated from an estimate of the content of the element in the specimen and from the amount of any carrier added, appears in the sixth columns of Tables 1, 2 and 3. In the case of cobalt in the Richardton aerolite, the isolation was essentially quantitative, and a chemical yield of 95 per cent, the same as obtained in the blank runs (Paper I), was assumed; from this the cobalt content of the gross meteorite material was calculated. The other figures for the weight fractions of Co in the specimens (second column, Table 3) and the weight fractions of Al_2O_3 (second column, Table 1) were taken from Tables 2 and 3 of Paper I. The weight fractions of BeO (second column, Table 2) are estimates. The granite and obsidian are assumed to contain 0·002% BeO, corresponding to 7 p.p.m. Be, the average value for acidic igneous rocks found by Sandell and Goldich (1943). The stone meteorites are assumed to contain 0·0003% BeO, corresponding to the mean Be abundance in chondrites of 1·2 p.p.m. adopted by

Table 1. Experimental data for Al^{26}

Category Specimen	Weight fraction Al_2O_3 (%)	Weight specimen (g)	Weight carrier Al_2O_3 (mg)	Weight sample counted (mg)	Fraction Al counted (%)	Net counting rate (count min^{-1})	$S(\theta)$	Sample specific activity (dis $min^{-1} g^{-1}$)	Specimen specific activity (dis $min^{-1} g^{-1}$)
Terrestrial									
Reagent	—	—	—	143·0	—	0·004 ± 0·010	0·72	0·1 ± 0·2	—
Ontario granite	14·3	179	—	200·0	0·78	−0·016 ± 0·010	0·67	−0·3 ± 0·2	−0·04 ± 0·03
California obsidian	13·7	100	—	97·4	0·71	0·011 ± 0·012	0·79	0·3 ± 0·4	0·05 ± 0·05
Aerolites									
Richardton	1·97	102	—	180·6	9·0	0·211 ± 0·008	0·68	4·2 ± 0·2	0·082 ± 0·003
Richardton (recycle)				181·6	9·0	0·161 ± 0·011	0·68	3·2 ± 0·2	0·063 ± 0·004
Plainview	2·67	200	—	91·5	1·72	0·063 ± 0·007	0·80	2·1 ± 0·2	0·056 ± 0·006
Plainview (recycle)				104·6	1·96	0·064 ± 0·010	0·77	1·9 ± 0·3	0·052 ± 0·008
Siderites									
Odessa	0	141	49·8	7·0	14·1	0·046 ± 0·008	1·02	15·7 ± 2·7	0·0055 ± 0·0010
Canyon Diablo	0	96	46·8	33·3	71	0·002 ± 0·007	1·03	0·1 ± 0·5	0·0001 ± 0·0002
Tektites									
Australite	13·5	70	—	257·8	2·7	0·015 ± 0·008	0·63	0·23 ± 0·12	0·031 ± 0·016
Australite (recycle)				313·7	3·3	0·029 ± 0·008	0·61	0·37 ± 0·10	0·050 ± 0·014
Bediasite	13·3	79	—	412·2	3·9	−0·001 ± 0·008	0·57	−0·01 ± 0·08	−0·001 ± 0·011
Moldavite	13·8	100	—	248·8	1·8	0·001 ± 0·007	0·64	0·02 ± 0·11	0·002 ± 0·015
Libyan glass	0·70	81	—	320·6	57	0·034 ± 0·008	0·60	0·43 ± 0·10	0·0030 ± 0·0007
Libyan glass (recycle)				129·4	23	0·016 ± 0·007	0·74	0·41 ± 0·18	0·0028 ± 0·0013

Table 2. Experimental data for Be^{10}

Category Specimen	Weight fraction BeO (%)	Weight specimen (g)	Weight carrier BeO (mg)	Weight sample counted (mg)	Fraction Be counted (%)	Net counting rate (count min^{-1})	$S(\theta)$	Sample specific activity (dis $min^{-1} g^{-1}$)	Specimen specific activity (dis $min^{-1} g^{-1}$)
Terrestrial									
Reagent	—	—	—	37·2	—	−0·015 ± 0·008	0·97	−0·9 ± 0·5	—
Ontario granite	0·002	179	38·6	4·0	9	0·008 ± 0·010	1·01	4·5 ± 5·6	0·0011 ± 0·0013
California obsidian	0·002	100	25·8	10·3	37	0·023 ± 0·008	1·02	5·0 ± 1·7	0·0014 ± 0·0005
Aerolites									
Richardton (recycle)	0·0003	102	38·6	68·0	(175)	0·311 ± 0·011	0·80	13·0 ± 0·5	(0·0087 ± 0·0003)
Richardton (2nd recycle)				26·1	67	0·154 ± 0·011	1·00	13·4 ± 1·0	0·0051 ± 0·0004
Plainview (recycle)	0·0003	400	38·6	70·7	(178)	0·228 ± 0·012	0·80	9·2 ± 0·5	(0·00162 ± 0·00009)
Siderites									
Odessa (recycle)	0	141	38·6	41·4	(107)	0·041 ± 0·010	0·95	2·4 ± 0·6	(0·00070 ± 0·00017)
Canyon Diablo	0	96	38·6	118·0	(306)	0·348 ± 0·013	0·62	10·8 ± 0·4	(0·0133 ± 0·0005)
Canyon Diablo (recycle)				76·1	(197)	−0·004 ± 0·008	0·77	−0·2 ± 0·3	(−0·0001 ± 0·0002)
Tektites									
Australite (recycle)	0·002	70	38·6	13·9	35	0·042 ± 0·008	1·02	6·7 ± 1·3	0·0038 ± 0·0007
Australite (2nd recycle)				8·6	22	0·011 ± 0·008	1·01	2·9 ± 2·1	0·0016 ± 0·0012
Bediasite (recycle)	0·002	79	38·6	10·6	26	0·048 ± 0·009	1·02	10·1 ± 1·9	0·0052 ± 0·0010
Moldavite	0·002	100	38·6	138·0	(340)	0·334 ± 0·012	0·57	9·7 ± 0·3	(0·0133 ± 0·0005)
Moldavite (2nd recycle)				86·6	(213)	0·087 ± 0·010	0·72	3·2 ± 0·4	(0·0028 ± 0·0003)
Moldavite (3rd recycle)				17·8	44	0·023 ± 0·009	1·01	2·9 ± 1·1	0·0012 ± 0·0005
Libyan glass (recycle)	0·0001	81	64·5	70·0	(108)	0·097 ± 0·009	0·80	3·9 ± 0·4	(0·0034 ± 0·0003)
Libyan glass (2nd recycle)				65·8	(102)	[0·144 ± 0·016]	0·81	[6·1 ± 0·7]	[0·0050 ± 0·0006]
Libyan glass (3rd recycle)				48·4	75	0·053 ± 0·009	0·92	2·7 ± 0·5	0·0022 ± 0·0004

Table 3. Experimental data for Co^{60}

Category Specimen	Weight fraction Co (%)	Equivalent CoS (%)	Weight specimen (g)	Weight sample counted (mg)	Fraction Co counted (%)	Net counting rate (count min^{-1})	$S(\theta)$	Sample specific activity (dis $min^{-1} g^{-1}$)	Specimen specific activity (dis $min^{-1} g^{-1}$)
Terrestrial									
Tech cobalt	—	—	—	146·6	—	0·003 ± 0·007	0·25	0·2 ± 0·5	—
Colorado cobalt	—	—	—	57·8	—	0·005 ± 0·006	0·55	0·4 ± 0·5	—
Ontario cobaltite	—	—	—	325·8	—	0·014 ± 0·010	0·12	0·9 ± 0·6	—
Aerolites									
Richardton	0·070	0·108	102	104·3	95	−0·001 ± 0·008	0·34	−0·1 ± 0·5	−0·0001 ± 0·0006
Plainview	0·050	0·077	200	79·0	51	0·057 ± 0·010	0·43	4·0 ± 0·7	0·0031 ± 0·0005
Plainview (recycle)				34·0	22	0·024 ± 0·010	0·74	2·3 ± 0·9	0·0018 ± 0·0007
Siderites									
Odessa	0·36	0·56	141	51·4	6·5	0·039 ± 0·006	0·59	3·1 ± 0·5	0·017 ± 0·003
Canyon Diablo	0·49	0·76	206	24·0	1·54	0·038 ± 0·013	0·86	4·4 ± 1·5	0·033 ± 0·011
Canyon Diablo (recycle)				31·2	2·00	0·045 ± 0·013	0·78	4·4 ± 1·3	0·033 ± 0·010
Canyon Diablo spheroids	0·95	1·47	5·0	41·0	56	0·011 ± 0·007	0·67	1·0 ± 0·6	0·014 ± 0·009

Table 4. Summary of experimental results

Category Specimen	Specimen specific Al^{26} activity (dis min^{-1} g^{-1})	Specimen specific Be^{10} activity (dis min^{-1} g^{-1})	Specimen specific Co^{60} activity (dis min^{-1} g^{-1})
Terrestrial			
Reagent aluminium	nil		
Reagent beryllium		nil	
Tech cobalt			nil
Colorado cobalt			nil
Ontario cobaltite			questionable
Ontario granite	<0·05	<0·0037	
California obsidian	<0·15	0·0014 ± 0·0005 *F*	
Aerolites			
Richardton	0·063 ± 0·004 *B*	0·0051 ± 0·0004 *B*	< 0·0012
Plainview	0·054 ± 0·005 *A*	0·00162 ± 0·00009 *D*	0·0018 ± 0·0007 *B*
Siderites			
Odessa	0·0055 ± 0·0010 *C*	0·00070 ± 0·00017 *C*	0·017 ± 0·003 *C*
Canyon Diablo	< 0·0006	<0·00049	0·033 ± 0·008 *A*
Canyon Diablo spheroids			<0·032
Tektites			
Australite	0·040 ± 0·011 *A*	0·0030 ± 0·0010 *B*	
Bediasite	< 0·022	0·0052 ± 0·0010 *F*	
Moldavite	< 0·032	0·0012 ± 0·0005 *E*	
Libyan glass	0·0030 ± 0·0007 *A*	0·0022 ± 0·0004 *B*	

Suess and Urey (1956). For the tektites and silica-glass, the Be : Al ratio is assumed to be the same as in the granite and obsidian.

It will be noted that in some cases the entry in the sixth column of Table 2 exceeds 100 per cent, for reasons mentioned earlier (Paper I). In these cases a value of 100 per cent was used in calculating the specimen specific Be^{10} activity; the latter is a lower limit because actual BeO yields are always less than 100 per cent. Apparent yields greater than 100 per cent and specific activities calculated therefrom are enclosed in parentheses.

Data for recycled samples are listed right after the previous measurements of the same sample. Because experience indicated that the first recycling of BeO almost invariably removed some activity, many of the BeO samples were not counted after the first time through the chemistry but were recycled immediately. In these cases the first value listed is the first recycle value, designated by "recycle". The designations "2nd recycle" and "3rd recycle" refer to sequential recyclings of the same BeO sample. Recycle sample weights can be larger than the previous sample weights because in many cases the samples mounted for counting were only portions of larger samples processed. The fraction of element counted (sixth column, Tables 1, 2 and 3) is thus not always the chemical yield and may be considerably lower.

The uncertainty index for each counting rate is the standard deviation (σ) calculated from counting statistics. These are the only uncertainties used in the calculation of the specific activities, because other sources of error are less important, and this gives the best way of deciding on whether two successive recycle values of the specific activity differ significantly from each other.

Table 4 gives, for each activity measured in each specimen, the best that can be said about the specimen specific activity from the data of Tables 1, 2 and 3. In case a net activity is less than twice its standard deviation (2σ), it is not regarded as significantly different from zero. Instead only an upper limit is given in Table 4. If the activity is zero or negative, the limit is taken to be 2σ. If the value is positive but less than 2σ, the limit is taken as the value plus 2σ. This "conservative 2σ criterion" assures that there is a 97·7 per cent probability that the true value does not exceed a limit so computed and that an accepted value is not actually zero.

In other cases, the entry in Table 4 is usually the last recycle measurement on the sample. In a few cases the next-to-last value is selected because the smaller sample size obtained upon recycling markedly reduced the accuracy, or an average of the last two values is adopted. A code is used to indicate the degree of reliability of the value. "*A*" means that the value has been established by recycling until the specific activity was constant, the difference in specific activity before and after recycling being less than one standard deviation of that difference. "*B*" refers to cases where the specific activity dropped on recycling, but to such a moderate extent as to indicate that the minimum value had very probably been reached. "*C*" designates single measurements after a degree of chemical processing which, according to other experience with similar materials, is almost certainly adequate for attaining the minimum activity level. "*D*" refers to single measurements in cases where experience indicated that a moderate decrease in specimen specific activity might occur on further purification. "*E*" indicates that a large decrease in specimen specific activity was observed upon the last recycling performed, suggesting strongly that the minimum level had still not been reached. Finally, "*F*" refers to single measurements following a degree of chemical processing which experience has indicated to be definitely inadequate. We regard the "*B*" and "*C*" values as essentially as reliable as the "*A*" values, even though lacking the rigorous "proof" of the latter.

The BeO from the Libyan desert silica-glass showed an increase of activity after the second chemical recycling. Investigation showed that the furnace in which the sample was ignited had been used just previously for igniting a Na^{22} source, and the BeO had presumably become contaminated with Na^{22} activity. The Be^{10} activity and specific activity figures in square brackets are therefore upper limits. This contamination should have been completely removed by the next recycling. A new furnace was used for subsequent ignitions.

4. Discussion

A. Terrestrial Materials and Possible Terrestrial Production

The reagent Al, Be and Co were all found to be completely inactive after the initial chemical procedures, even without recycling. The same chemicals were used for carriers when required.

The Al_2O_3 isolated from granite and obsidian were also inactive. However, the high alumina content of these materials makes it difficult to establish a very low limit to the Al^{26} activity level in terrestrial silicates. Somewhat lower specimen specific activities were obtained with the bediasite and moldavite tektites, and these constitute the best experimental evidence that the terrestrial production of Al^{26} is not significant.

In the case of Be^{10}, the BeO from granite was inactive, but unfortunately here the low chemical yield prevented the establishment of a low specimen specific activity limit. The only measurement on the obsidian sample gave a net activity 2·9 times its standard deviation, which is statistically significant; however, this sample was not recycled, and experience with tektites suggests that contaminating activities, probably members of the uranium and thorium families, are not completely removed by the initial chemical procedure for materials of this general composition.

None of the terrestrial cobalt samples showed a statistically significant activity. The CoS from the Ontario cobaltite had a net activity 1·4 times its standard deviation, suggestive of the presence of Co^{60}. Although this sample was exposed at the surface until a few months before measurement in a rather high magnetic latitude (58° N), the elevation (~320 m) was not great enough to lead to expectation of a particularly high cosmic-ray slow-neutron flux.

Most of the ionization produced by cosmic-rays at ground level is due to secondary particles, muons and electrons, which are very ineffective in inducing nuclear reactions. Gamma radiation is also prominent among the secondaries, but photonuclear reaction cross-sections are rather small. Only the nucleonic constituents, protons and neutrons, are relatively effective in producing reactions, but their concentrations are very low. Moreover, only a small fraction of them have sufficient energy to produce spallation-type reactions. Accordingly, one would expect that the ability of cosmic radiation at low or moderate altitudes to produce such nuclides as Al^{26} and Be^{10} would be very much less than that of the radiation outside of the earth's atmosphere. Consideration of the levels observed in stone meteorites indicates that the actual levels in favourably situated terrestrial materials at low or moderate altitudes should be much lower than our experimental limits. Davis (1955) observed in CCl_4 exposed to cosmic rays at an altitude of 4300 m a saturation A^{37} activity, induced principally by the nucleonic component via Cl^{37} (p,n), of ~0·000011 dis min^{-1} $(g\ CCl_4)^{-1}$. At sea level the effect was $<0{\cdot}0000006$ dis min^{-1} $(g\ CCl_4)^{-1}$. At the highest altitude of any of our specimens, 1800 m, the intensity of the nucleonic component is about one-fifth of its value at 4300 m. The production rate of Al^{26} in silicates should not be grossly different from that of A^{37} in chlorine, and the Be^{10} production rate should be considerably lower. This likewise leads to the conclusion that saturation Al^{26} and Be^{10} activities from cosmic rays at low or moderate altitudes should be negligible. Moreover, it is highly unlikely that a terrestrial object now found on the earth's surface could have been there for more than a small fraction of the half-life of Al^{26} or Be^{10}; most of its existence would have been spent 1 m or more underground very effectively shielded from the cosmic radiation.

In the ground, and also in meteorites and tektites, some Be^{10} production might

be expected from the following "simple" nuclear reactions caused by radiations associated with natural radioelements:

(a)	$(92{\cdot}5\%)Li^{7}(\alpha,p)Be^{10}$	$Q = -2{\cdot}57$ MeV	
(b)	$(100\%)Be^{9}(n,\gamma)Be^{10}$	$Q = +6{\cdot}81$ MeV	$\sigma_{th} = 0{\cdot}010$ barn
(c)	$(18{\cdot}8\%)B^{10}(n,p)Be^{10}$	$Q = +0{\cdot}23$ MeV	$\sigma_{th} < 0{\cdot}2$ barn
(d)	$(81{\cdot}2\%)B^{11}(\gamma,p)Be^{10}$	$Q = -11{\cdot}24$ MeV	
(e)	$(98{\cdot}9\%)C^{12}(n,He^{3})Be^{10}$	$Q = -19{\cdot}47$ MeV	
(f)	$(1{\cdot}1\%)C^{13}(n,\alpha)Be^{10}$	$Q = -3{\cdot}85$ MeV	

However, reaction (a) should be important only in materials containing moderate concentrations of both lithium and uranium or thorium; (b) and (c) have too small cross-sections to be important with thermal neutrons, and with fast neutrons yields could be appreciable only in Be- or B-rich materials associated with abundant U- or Th-rich materials; (d) and (e) have much too high thresholds; and (f) combines a moderately high threshold with a low target isotopic abundance. Accordingly, in ordinary rocks, as well as in meteorites and tektites, production of Be^{10} in this manner should be negligible. Similarly, the following "simple" reactions might lead to Al^{26}:

(g)	$(100\%)Na^{23}(\alpha,n)Al^{26}$	$Q = -2{\cdot}95$ MeV
(h)	$(100\%)Al^{27}(\gamma,n)Al^{26}$	$Q = -13{\cdot}04$ MeV
(i)	$(100\%)Al^{27}(n,2n)Al^{26}$	$Q = -13{\cdot}04$ MeV
(j)	$(92{\cdot}17\%)Si^{28}(n,H^{3})Al^{26}$	$Q = -16{\cdot}15$ MeV

However, reactions (*h*), (*i*) and (*j*) have too high thresholds on grounds of energetics, and the threshold of (*g*) as determined by electrostatic barrier and momentum conservation considerations is about 7 MeV. Hence, except conceivably in materials rich in sodium and thorium or uranium, Al^{26} production by non-cosmic radiations should be utterly insignificant.

The theoretical considerations are a stronger basis than the experimental evidence for our conclusion that any Al^{26} or Be^{10} observed by our techniques in any of the materials examined by us must have been produced by cosmic radiation in space.

As regards terrestrial production of Co^{60} via $Co^{59}(n,\gamma)$, calculations similar to those made by Davis and Schaeffer (1955) for Cl^{36} lead one to expect a Co^{60} specific activity of $\sim 0{\cdot}5$ dis min^{-1} (g CoS)$^{-1}$ in cobalt saturated at 2000 m in mean magnetic latitude and of $\sim 0{\cdot}1$ dis min^{-1} (g CoS)$^{-1}$ at sea level. The neutron production in ordinary rocks as a result of their radioelement content, $\sim 0{\cdot}00005$ neutron $min^{-1}\,g^{-1}$ (Pine and Morrison, 1952), is even less than the production by cosmic rays at sea level. Thus the expected levels of natural Co^{60} activity in any of our samples are at or below the sensitivity limit of our present technique. In samples from several thousand metres altitude, however, cosmic-ray-induced Co^{60} should be readily measurable (Davis and Schaeffer, 1955).

B. Aerolites (Stone Meteorites)

The Al^{26} result on the Plainview chondrite, which meets our rigorous specifications for an "*A*" classification, establishes without doubt the occurrence of long-lived cosmic-ray-induced radioactivity in meteorites. The final "*B*" value for the

Richardton chondrite is regarded as equally reliable. Since these values differ by little over one standard deviation of their difference, they probably define the general level of this nuclide in moderate-sized stone meteorites. It will be recalled that, according to PRIOR and HEY (1953), approximately 100 kg and 700 kg or Richardton and Plainview meteorites, respectively, have been recovered. The larger size of the latter may have resulted in more shielding and may account for the slightly lower value. However, the radius of a spherical aerolite weighing as much as 2000 kg would only be about 50 cm, so most of its substance would be in the zone of relatively high induced reaction rate (MARTIN, 1953).

The final "*B*" value for the Be^{10} measurement in the Richardton chondrite, though possibly still a little high, is certainly reliable enough to establish the occurrence and general level of this nuclide in moderate-sized stone meteorites. The small ratio ($\sim$1 : 12) between the Be^{10} and Al^{26} activities in this object corresponds to the facts that Be^{10} can be produced only by spallation-type reactions (principally from O^{16} by $(p,5p2n)$ and $(p,3p\alpha)$, with smaller contributions from Mg and Si spallation and Fe fragmentation), whereas Al^{26} can be produced more effectively by "simple" reactions on abundant adjacent elements ($Na^{23}(\alpha,n)$, $Mg^{24}(\alpha,pn)$, $Mg^{26}(p,n)$, $Si^{28}(p,2pn)$, $Si^{29}(p,\alpha)$ and $Si^{30}(p,n\alpha)$), as well as by high-energy processes [$Si^{28}(\pi^-,2n)$ and spallation of heavier elements].

The "*D*" value on Be^{10} in the Plainview chondrite may be regarded as an upper limit. Therefore, the Be^{10} : Al^{26} ratio in Plainview ($<$1 : 30) is distinctly lower than in Richardton. There are two possible explanations for this: (1) the Plainview specimen may have been farther from the surface of the meteoroid, so that the primaries and high-energy secondaries, by which Be^{10} is produced, may have been largely removed from the flux, whereas the lower-energy secondaries, which are still influential in producing Al^{26}, may still have been present. This would correspond to the probable larger size of the Plainview object. (2) The Plainview specimen may have been exposed to the cosmic radiation for only $\sim 10^6$ years, in which case the Al^{26} activity would be over half-way to its saturation level but the Be^{10} would be less than one-quarter of the way to its saturation level; while the Richardton specimen may have been exposed for $\sim 10^7$ years or longer, so that both nuclides would be in secular equilibrium with the flux. That the meteorites falling on earth are fairly "young" as small bodies follows from the calculations of PIOTROWSKI (1952) that collisions of asteroids and intermediate-sized bodies are taking place at appreciable rates in the solar system even at the present time, and of ÖPIK (1951) that objects in orbits crossing that of the earth have expectation lifetimes of only $\sim 10^8$ years before collision with the earth. FIREMAN and SCHWARZER (1957) have determined cosmic-ray exposure ages from the He^3/H^3 ratio of 900 and 1700 million years for two iron meteorites. EBERT and WÄNKE (1957) have obtained an exposure time of 920 million years for an iron meteorite from its neon content and a calculated production rate. BEGEMANN, GEISS and HESS (1957) have determined a He^3/H^3 age of 260 million years for an achondrite. REED and TURKEVICH (1957) have calculated He^3 ages, using an assumed production rate, of eleven chondrites, ranging from 4·7 to 112 million years. The more fragile stone meteorites might be expected to have shorter average lifetimes than irons. Thus several per cent of the meteorites falling to

earth should be unsaturated in Be^{10} activity. Which of these two explanations is correct here can be settled by studies of the depth dependence of activity of each nuclide, measurements of radionuclides having different lifetimes, and measurements of stable nuclear reaction products.

The "*B*" value for Co^{60} in the Plainview chondrite may be regarded as definite evidence for the occurrence of this nuclide, in view of its absence from the Richardton specimen following even less rigorous chemistry. Since the former object fell at least 40 years ago, all Co^{60} produced directly by cosmic rays in space has decayed away, leaving two possible explanations for the present content of Co^{60} (1) presence of the long-lived Fe^{60}, produced by cosmic rays, with which Co^{60} would be in secular equilibrium; (2) production of Co^{60} by neutron capture in the cobalt of the meteorite in the last few years while the object has been on earth. If the former explanation were correct, one would expect Fe^{60} and Co^{60} in the Richardton meteorite, possibly at an even higher level, because, like Be^{10}, Fe^{60} should be produced only by high-energy processes [particularly $Ni^{62}(p,3p)$, $Ni^{64}(p,p\alpha)$ and $Ni^{62}(\pi^-,pn)$]. The absence of Co^{60} and hence of Fe^{60} in Richardton argues against the presence of Fe^{60} in Plainview, and indicates that the latter's Co^{60} must be the result of terrestrial neutron capture. Whether the neutrons were from ground-level cosmic radiation, from naturally radioactive substances stored near the specimen, or from artificial neutron sources cannot be stated without more detailed information on the history of the specimen.

C. Siderites (Iron Meteorites)

Very low limits to the possible presence of Al^{26} and Be^{10} were set in the Canyon Diablo meteorite specimen. Therefore the "*C*" results on the Odessa siderite, obtained with the identical chemistry, can be regarded as reliable. The Be^{10} : Al^{26} ratio of $\sim 1 : 8$ is consistent with the fact that Be is considerably further than Al from the principal target elements, Fe and Ni; that the ratio is not still smaller may be due to a substantial augmentation of the spallation yield of Be^{10} by the "fragmentation" process, in which rather substantial nuclear aggregates are expelled in high-energy nuclear collisions. Each of these nuclides is present in the Odessa iron meteorite at roughly one-tenth of its level in the Richardton chondrite. This seems consistent with the much greater distance between target and product nuclides in metal as compared to silicate meteorites.

Both the internal comparison and the comparisons with the Richardton chondrite suggest that the Odessa meteorite, and accordingly also its associated craters, are probably not older than about one Al^{26} half-life, or less than about a million years. Fairly accurate dating of the fall of this object should be possible through measurements of shorter-lived nuclides, particularly $Cl^{36}(H = 3{\cdot}1 \times 10^5$ years), $Ca^{41}(1{\cdot}1 \times 10^5$ years) and $C^{14}(5{\cdot}7 \times 10^3$ years).

The absence of Al^{26} and Be^{10} in the Canyon Diablo specimen may be explained in two ways: (1) a terrestrial age of at least several million years, during which these two nuclides have decayed to below the levels of detection; or (2) a shielded position for the particular specimen examined deep inside a large mass. The first explanation is made unlikely by the apparent geological youthfulness of the associated Meteor Crater. The latter is much better preserved than the main

Odessa crater, and is, therefore, almost certainly younger, since both lie in similar arid terrains. The most thorough geological estimation of the age, by Blackwelder (1932), places it as probably between 40,000 and 75,000 years. Therefore, shielding is almost certainly responsible. Estimates of the mass which produced the crater vary widely. Although Wylie (1943) and Rinehart (1950) assign masses $\sim 10^4$ tons, La Paz (1943) and Rostoker (1953) concur with Öpik (1936), whose estimate was ~4,000,000 tons. The latter mass corresponds to a radius of ~50 m, and even the smaller masses correspond to radii of ~7 m. The majority of the substance of the meteoroid would therefore be adequately shielded from the cosmic radiation by over 1 m of iron, and the probability that our specimen was so situated is high.

Thus the present measurements do not contribute to the age of the Meteor Crater. However, He^3, A^{36} and A^{38} produced by cosmic rays have been observed in a specimen of Canyon Diablo iron (Gentner and Zähringer, 1957), although at a level much lower than in other siderites. It might be expected that the recoverable fragments of such a giant meteorite would come largely from the back side of the object as it ploughed into the earth, with the front and central material and the compressed rock absorbing most of the kinetic energy of the entire object as heat and exploding, throwing the cushioned rear-end material out. Thus among these fragments some might have been in the zone of fairly high cosmic-irradiation intensity. Examination of a number of Canyon Diablo specimens for induced radionuclides will almost certainly provide a basis for determining the age of the Meteor Crater. It might also yield information on the size of the meteoroid, and contribute to determining the status of the existence of two major varieties of Canyon Diablo irons, oxidation-resistant and chlorine-rich or oxidizing (Fairchild, 1907), and of the atypical Canyon Diablo irons discovered by Nininger and referred to by him as Canyon Diablo No. 2, No. 3, and No. 4 (1952, p. 234; 1956, p. 64).

Moderately high and similar quantities of Co^{60} were found in both Canyon Diablo and Odessa specimens. Whatever the reason for the absence of Al^{26} and Be^{10} in the Canyon Diablo specimen, Fe^{60} should be absent for the same reason. Therefore the Co^{60} in this material must be a result of recent terrestrial neutron capture. The Co^{60} in the Odessa specimen may be partially or wholly due to the same cause. However, some or all of it may be due to the presence of Fe^{60}; this could be established by "milking" of cobalt from the separated iron of the meteorite after it has stood for several years allowing some Co^{60} to grow in again.

The absence of a definitely positive Co^{60} result on the Canyon Diablo spheroids suggests that neutron activation of Co^{59} at this locality is probably not significant. The spheroids should have been well exposed to the cosmic-ray neutrons up to the time of their collection, which was not over 2 years before measurement and they were stored at nearly the same altitude until a few months before measurement. The important comparison here is between the sample specific activities rather than the specimen specific activities (Table 3). Although the net effect here of 1·6 times its standard deviation is suggestive of Co^{60} activity, we suspect that the Co^{60} activities which we have observed are less reflective of the neutron fluxes at the places of discovery than of those at the places of subsequent storage.

D. Tektites and Silica-Glass

The australite specimen showed definite Al^{26} activity, essentially constant through a recycle, at a level similar to that in the stone meteorites. In view of the absence of this nuclide in the bediasite and moldavite specimens, and the above-mentioned (Section 4 A) theoretical arguments against terrestrial production of Al^{26} at levels comparable to those in meteorites, we must conclude that the australites have spent about a million years or longer exposed to the cosmic radiation outside of the earth's atmosphere.

This result indicates, with a high degree of probability, that the tektites are glass meteorites. This is in accord with the opinions of several prominent investigators, including F. E. Suess (1900), Lacroix (1932), Fenner (1935, 1938, 1949), Paneth (1940, 1949), Baker (1944), H. E. Suess (1951), Barnes (1951), Stair (1956) and Cassidy (1956), but in contrast with those of Spencer (1933, 1937), Krinov (1955), Urey (1955, 1957), and formerly Barnes (1940). The latter group have given strong arguments against a meteoritic nature of tektites and in favour of an origin from terrestrial materials, although perhaps as the result of collisions of moderate-sized extra-terrestrial objects with the earth. It must be admitted that positive evidence for a meteoritic origin of tektites has hitherto been absent. Even if the present results are accepted as conclusive evidence for such an origin, the explanation of their nature and occurrence poses difficult problems.

The Al^{26} content of the Libyan Desert silica-glass, likewise an "*A*" result, is only about one-twentieth of that of the chondrites and australites, but is still far above the level expected from terrestrial production at an altitude of 400 m. Here too a cosmic origin is indicated, as has been favoured by those favouring such an origin for tektites and by Oakley (1952) and not excluded by Spencer (1937). Thus it must be considered a variety of tektite; its differences from the other tektites (Spencer, 1933, 1939; Barnes, 1940; Stair, 1955) can be regarded as of degree rather than of kind.

Be^{10} also appears to be definitely present in the australite and Libyan glass. In the former case the activity decreased on recycling by only 1·6 times the standard deviation of the decrease, and the adopted value, a sort of average of the last two measurements, is classified as "*B*". The measurements of the Libyan glass were unfortunately interfered with by the laboratory contamination of the second recycle sample, but if most of the drop between the first and third recycle values occurred on the second recycling, as would be expected, the drop between the second and third recycle values would most probably have been less than its standard deviation, meriting an "*A*" classification rather than the "*B*" rating actually assigned. The activity levels are again comparable to those in the stone meteorites, strengthening the conclusion about the meteoritic nature of tektites including the Libyan glass.

The residual activity in the bediasite and moldavite BeO samples could well be a result of insufficient chemical processing, as in the case of the obsidian. The bediasites (Barnes, 1940) and moldavites (Suess, 1900) are associated with Upper Eocene and Middle Miocene deposits, respectively, and have therefore been on earth about 50 and 20 million years. One would expect Be^{10}, like Al^{26}, to be

absent. Demonstration that this is the case would be important in confirming the significance of the australite and Libyan glass results.

The levels or limits of Al^{26} and Be^{10} activities give information about the times of fall of the various tektite groups. The australites are probably not much older than one-half of one Al^{26} half-life, or $<0{\cdot}5$ million years old. The Libyan glass Al^{26} and Be^{10} specific activity levels are respectively about 0·05 and 0·4 times the levels in the Richardton chondrite; both observations are consistent with an age of $\sim 3{\cdot}5$ million years. The bediasite and moldavite Al^{26} levels give age limits of $>1{\cdot}2$ and $>0{\cdot}8$ million years respectively. The moldavite Be^{10} result sets an upper limit to the specimen specific activity (0·0021 dis min^{-1} g^{-1}) corresponding to an age limit of >3 million years. These ages and limits are consistent with the occurrence and superficial erosion of the various groups of objects. The exact values given are, of course, based on the assumption that our chondrite results are indicative of the tektite activities at their time of fall. This assumption may be incorrect; the calculations have been carried out mainly as an illustration of the eventual possibilities of this dating method.

E. Past Cosmic-ray Intensity

The above discussions of the activity levels of induced radioactivities in stone and iron meteorites, and of the terrestrial ages of tektites, are underlain by the tacit assumption that the cosmic radiation intensity over long periods in the past has been essentially constant at approximately its present value. The same assumption underlies the calculation of cosmic-ray exposure ages of meteorites from their contents of stable nuclear reaction products. Studies of the radioactive products should yield information on this point, the equilibrium level of each nuclide serving as an index to the mean intensity over a period of time in the past approximating the mean lifetime of the nuclide (Singer, 1953; Gentner and Zähringer, 1957; Sheline and Hooper, 1957; Fireman and Schwarzer, 1957). The radionuclides possess the merit that the exposure age need not be known as long as it can be presumed (or shown from observations on longer-lived or stable nuclides) to be long relative to the half-lives involved.

We have seen that the Be^{10} and Al^{26} results in stone and iron meteorites appear to be mutually consistent and in line with the theoretical expectations (Paper I). From this we might be justified in concluding that the average cosmic-ray flux has not changed grossly over the past few million years. However, because of the lack of quantitative data on the relation between the production rates of various nuclides and the cosmic-ray intensity and, more seriously, because of the strong dependence of the production rates on size of meteorite and depth of specimen beneath the pre-atmospheric surface (Paneth, Reasbeck and Mayne, 1953; Gentner and Zähringer, 1955, 1957; Fireman and Schwarzer, 1957; Ebert and Wänke, 1957), a precise statement on this point cannot be made now.

A promising approach to this problem would be to measure the activity levels of several different radionuclides in the same specimen, and to compare their ratios with the pattern determined by bombardments with high-energy accelerators and by observations on stable reaction products. The available data on He, Ne and A products indicates that in iron meteorites the element and isotope ratios

are fairly constant in spite of large variations in their concentration levels (Reasbeck and Mayne, 1955; Gentner and Zähringer, 1955, 1957; Ebert and Wänke, 1957). Goel (1958) has given arguments, based on the characteristics of "stars" produced in nuclear emulsions by cosmic rays, for generalizing that the production rate pattern should be practically the same for all depths in all iron meteorites. If so, this would make uncertainty in the positions of the individual specimens of little consequence.

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